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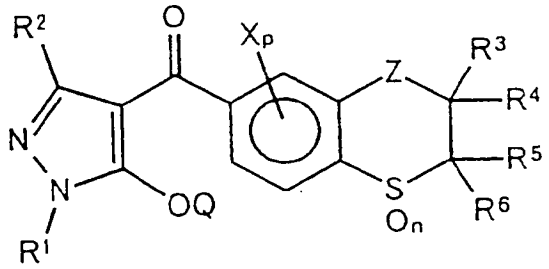
(71) Applicant: **IDEMITSU KOSAN COMPANY LIMITED**
Tokyo 100 (JP)

(72) Inventors:
- **SHIBATA, Mitsuru**
  Sodegaura-shi Chiba-ken 299-02 (JP)
- **NASUNO, Ichiro**
  Sodegaura-shi Chiba-ken 299-02 (JP)
- **NAKAMURA, Kazufumi**
  Sodegaura-shi Chiba-ken 299-02 (JP)
- **KOIKE, Kazuyoshi**
  Sodegaura-shi Chiba-ken 299-02 (JP)
- **YOSHIKAWA, Misako**
  Sodegaura-shi Chiba-ken 299-02 (JP)

(74) Representative: **Türk, Gille, Hrabal, Leifert**
Brucknerstrasse 20
40593 Düsseldorf (DE)

(54) HERBICIDAL COMPOSITION

(57) Provided by the present invention is a herbicide composition containing, as active ingredients, a pyrazole derivative of the general formula (I)



(I)

and at least one herbicide compound selected from the group consisting of chloroacetamide-containing herbicides, imidazoline-containing herbicides, atrazine, cyanazine, metribuzin, linuron, metbenzurone, bentazone, dicamba, chlopyralid, 2,4-D, bromoxynil, pendimethalin, nicosulfuron, rimsulfuron, primisulfuron and pyridate. The herbicide composition of the present invention can control a broad rage of gramineous weeds and broad-leaved weeds at a very low dosage without damaging corn and other crops.

EP 0 768 033 A1

Description

TECHNICAL FIELD

The present invention relates to a herbicide composition containing pyrazole derivative.

TECHNICAL BACKGROUND

Herbicides are very important chemicals for saving weed-controlling labors and improving the yield of agricultural and horticultural crops and have been therefore aggressively studied and developed for many years, and a diversity of herbicides have now been put to practical use. However, it cannot be said that herbicides which have been so far developed have broad herbicidal spectra, and at present there are an increasing number of weeds which are difficult to control. Herbicides having broad herbicidal spectra are therefore desired. For overcoming the environmental pollution problem of conventional herbicides, further, there are desired herbicides which are effective at a further decreased dosage.

The present inventors already found that a variety of pyrazole derivatives in which a thiochroman ring is bonded to a pyrazole ring through a carbonyl group is free from damaging gramineous upland crops but can control gramineous weeds and broad-leaved weeds together at a low dosage by any one of soil treatment and foliar treatment, and filed patent applications directed to these pyrazole derivatives and herbicides containing these as active ingredients. Of these patent applications, some have been already laid-open as International Laid-open Patent Publications Nos. WO95/04054, WO93/18031, WO94/01431 and WO95/13275. Japanese Patent Applications Nos. 6-237981, 7-80059 and 7-158842 have not yet been laid-open.

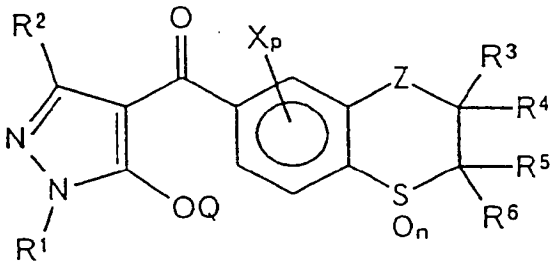
Those pyrazole derivatives disclosed in the above International Laid-open Patent Publications and Japanese Patent Applications can control gramineous weeds and broad-leaved weeds together at a low dosage by any one of soil treatment and foliar treatment, while there is desired a herbicide which can control a broad range of gramineous weeds and broad-leaved weeds at a further decreased dosage.

It is therefore an object of the present invention to provide herbicide compositions which can control a broad range of gramineous weeds and broad-leaved weeds at a very low dosage without damaging crops such as corn, etc., by using the pyrazole derivatives disclosed in the International Laid-open Patent Publications and the above Japanese Patent Applications in combination with other herbicide compounds.

The present inventors have made diligent studies to achieve the above object, and as a result have found that when a pyrazole derivative of the following general formula (I) described in the above International Laid-open Patent Publications and Japanese Patent Applications (to be sometimes referred to as "pyrazole derivative (I)" hereinafter) and a specific herbicide compound are used in combination, a broad range of gramineous weeds and broad-leaved weeds can be controlled at a very low dosage by synergistic effects of the pyrazole derivative (I) and the said specific herbicide compound without damaging crops such as corn, etc. The present invention has been accordingly completed.

DISCLOSURE OF THE INVENTION

The gist of the present invention is a herbicide composition containing, as active ingredients, a pyrazole derivative of the general formula (I),



(I)

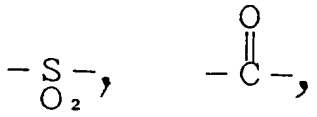
{wherein:

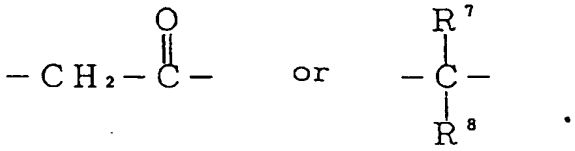
R^1 is a C_1~C_4 alkyl group, a C_2~C_4 alkenyl group or a C_2~C_4 haloalkeny group,
R^2 is a hydrogen atom, a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group or a C_2~C_4 alkoxyalkyl group,
X is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_2~C_4 alkoxyalkyl group, a halogen atom, a C_1~C_4 alkoxy

group or a C_1~C_4 haloalkoxy group,
p is an integer of 0, 1 or 2,
each of R^3, R^4, R^5 and R^6 is independently a hydrogen atom, a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group or a C_2~C_4 alkoxyalkyl group,
n is an integer of 0, 1 or 2,
Q is a hydrogen atom or a group of A-B,

[in which

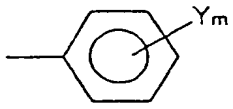
A is a group of





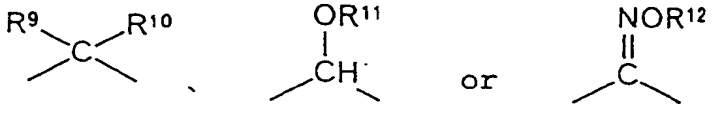
(in which each of R^7 and R^8 is independently a hydrogen atom or a C_1~C_4 alkyl group), and

B is a C_1~C_{12} alkyl group, a C_3~C_{10} cycloalkyl group or a group of



(in which Y is a C_1~C_4 alkyl group, a C_1~C_4 alkoxy group, a C_1~C_4 haloalkyl group, a nitro group or a halogen atom, and

m is an integer of 0 or 1 - 3)], and
Z is



[in which

R^9 is a hydrogen atom, a C_1~C_4 alkyl group or a C_1~C_4 haloalkyl group,

R^{10} is a hydrogen atom, a C_1~C_4 alkyl group, a C_2~C_4 alkenyl group or a C_2~C_4 alkynyl group,

R^{11} is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_3~C_6 cycloalkyl group, a C_3~C_6 alkenylalkyl group, a C_3~C_6 alkynylalkyl group or a C_3~C_6 haloalkenylalkyl group,

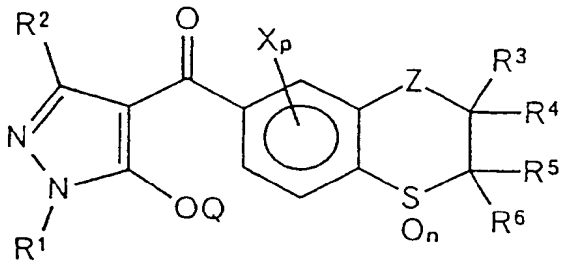
R^{12} is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_3~C_6 cycloalkyl group, a C_3~C_6 alkenylalkyl group, a C_3~C_6 alkynylalkyl group or a C_3~C_6 haloalkenylalkyl group]},

or a salt thereof; and

at least one herbicide compound selected from the group consisting of chloroacetamide-containing herbicides such as alachlor, metolachlor, acetochlor and dimethenamide, etc., imidazoline-containing herbicides such as imazetapyr, etc., atrazine, cyanazine, metribuzin, linuron, metbenzurone, bentazone, dicamba, chlopyralid, 2,4-D, bromoxynil, pendimethalin, nicosulfuron, rimsulfuron, primisulfuron and pyridate.

PREFERRED EMBODIMENTS OF THE INVENTION

The pyrazole derivative as a first active ingredient of the herbicide composition of the present invention has the following general formula (I).



(I)

The herbicide composition of the present invention contains, as an active ingredient, at least one of pyrazole derivatives of the above general formula (I).

In the general formula (I), R^1 is a C_1~C_4 alkyl group, a C_2~C_4 alkenyl group or a C_2~C_4 haloalkenyl group, and preferred is a C_1~C_4 alkyl group. Specific examples of the C_1~C_4 alkyl group include methyl, ethyl, propyl and butyl, and the propyl and the butyl may be any one of linear, cyclic and branched ones. The C_1~C_4 alkyl group is preferably methyl or ethyl. Specific examples of the C_2~C_4 alkenyl group include $-CH=CH_2$, $-CH_2-CH=_{CH2}$ and $-CH=CH-CH=CH_2$. The C_2~C_4 haloalkeny group includes those in which at least one hydrogen atom of the above C_2~C_4 alkenyl group is replaced with a halogen atom (e.g., a chlorine atom, a fluorine atom, a bromine atom or an iodine atom).

In the general formula (I), R^2 is a hydrogen atom, a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group or a C_2~C_4 alkoxyalkyl group, and preferred is a hydrogen atom or a C_1~C_4 alkyl group. Specific examples of the C_1~C_4 alkyl group are as described concerning R^1, and methyl is preferred. The C_1~C_4 haloalkyl group includes those in which at least one hydrogen atom of the C_1~C_4 alkyl group is replaced with a halogen atom (e.g., chlorine atom, a fluorine atom, a bromine atom or an iodine atom), and examples thereof include $-CF_3$, $-C_2F_5$, $-C_2H_4F$, $-CH_2Cl$, $-CHF_2$, $-CCl_3$, $-C_2H_3Cl_2$ and $-C_2H_3F_2$. Specific examples of the C_2~C_4 alkoxyalkyl group include $-CH_2-OCH_3$, $-CH_2-OC_2H_5$, $-CH_2-OC_3H_7$, $-CH(CH_3)OCH_3$, $-CH(CH_3)OC_2H_5$, $-CH_2CH_3OCH_3$ and $-CH_2CH_2OC_2H_5$.

In the general formula (I), X is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_2~C_4 alkoxyalkyl group, a halogen atom, a C_1~C_4 alkoxy group or a C_1~C_4 haloalkoxy group, and preferred is a C_1~C_4 alkyl group or a halogen atom. Specific examples of the C_1~C_4 alkyl group, the C_1~C_4 haloalkyl group and the C_2~C_4 alkoxyalkyl group are as described concerning R^1 or R^2. Specific examples of the C_1~C_4 alkyl group preferably include methyl or ethyl. Specific examples of the halogen atom include a chlorine atom, a fluorine atom, a bromine atom and an iodine atom, and a chlorine atom is preferred. Specific examples of the C_1~C_4 alkoxy group include methoxy, ethoxy, propoxy and butoxy, and the propoxy and the butoxy may be linear, cyclic or branched ones. The C_1~C_4 haloalkoxy group include those in which at least one hydrogen atom of the C_1~C_4 alkoxy group is replaced with a halogen atom (e.g., a chlorine atom, a fluorine atom, a bromine atom or an iodine atom), and examples thereof include $-OCF_3$, $-OC_2F_5$, $-OC_2H_4F$, $-OC_2H_4Cl$, $-OCHF_2$, $-OCH_2F$, $-OCCl_3$, $-OC_2H_3Cl_2$ and $-OC_2H_3F_2$.

In the general formula (I), p is the number of substituent(s) X, and it is an integer of 0, 1 or 2. When p is 1 or 2, the position(s) of the substituent(s) X is/are the 5-position and/or 8-position on the thiochroman ring.

In the general formula (I), each of R^3, R^4, R^5 and R^6 is independently a hydrogen atom, a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group or a C_2~C_4 alkoxyalkyl group. Each of these is preferably independently a hydrogen atom or a

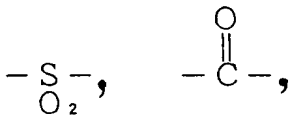
C_1~C_4 alkyl group. Specific examples of the C_1~C_4 alkyl group, the C_1~C_4 haloalkyl group and the C_2~C_4 alkoxyalkyl group are as described concerning R^1 or R^2.

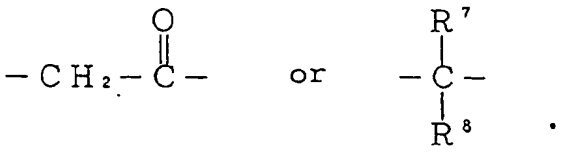
In the general formula (I), n is the number of oxygen atom(s) bonding to the sulfur atom of the thiochroman ring, and it is an integer of 0 (sulfide), 1 (sulfoxide) or 2 (sulfone) and is preferably 2 (sulfone).

In the general formula (I), Q is a hydrogen atom or a group of -A-B.

In the definition of Q,

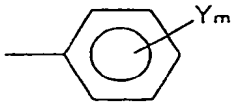
A is a group of





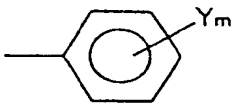
In the definition of A, each of R^7 and R^8 is independently a hydrogen atom or a C_1~C_4 alkyl group, and each is preferably a hydrogen atom. Specific examples of the C_1~C_4 alkyl group are as described concerning R^1.

In the definition of Q, B is a C_1~C_{12} alkyl group, a C_3~$_{10}$ cycloalkyl group or a group of



Specific examples of the C_1~C_{12} alkyl group includes those described as specific examples of the C_1~C_4 alkyl group for R^1 and others such as pentyl, hexyl, heptyl, octyl, nonyl, decanyl, undecanyl and dodecanyl. Those which have at least 3 carbon atoms may be linear or branched. Preferred is a C_1~C_8 alkyl group. Specific examples of the C_3~C_{10} alkyl group include cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl and cycloheptyl, and cyclohexyl is preferred.

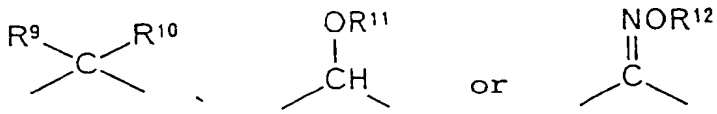
In the group of



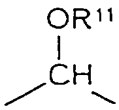
which is one embodiment of B, Y is a C_1~C_4 alkyl group, a C_1~C_4 alkoxy group, a C_1~C_4 haloalkyl group, a nitro group or a halogen atom, and preferred is a C_1~C_4 alkyl group, a C_1~C_4 alkoxy group, a nitro group or a halogen atom. Specific examples of the C_1~C_4 alkyl group, the C_1~C_4 alkoxy group, the C_1~C_4 haloalky group and the halogen atom are as described concerning R^1, R^2 or X. The C_1~C_4 alkyl group is specifically preferably methyl. The C_1~C_4 alkoxy group is specifically preferably methoxy. The halogen atom is specifically preferably a chorine or fluorine atom.

m is the number of substituent(s) Y, and it is an integer of 0 or 1 to 3, preferably 0, 1 or 2.

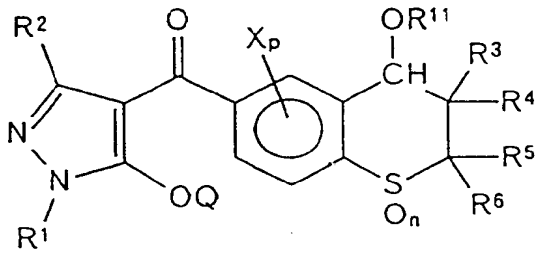
In the general formula (I), Z is



The pyrazole derivative (I) of the present invention can be classified into three kinds depending upon embodiments of Z. That is, the pyrazole derivative in which Z is

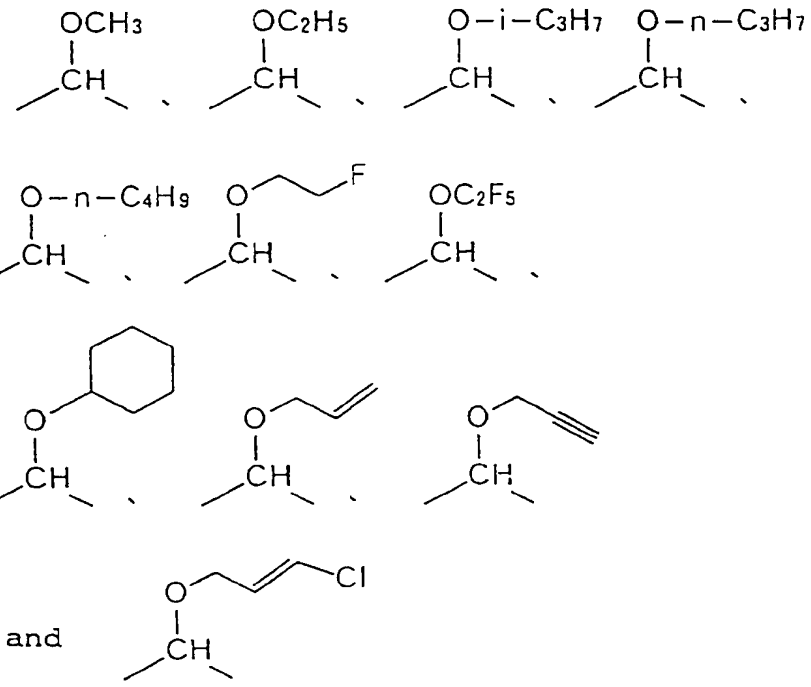


has the following general formula (Ia).

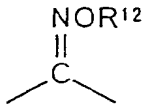


(Ia)

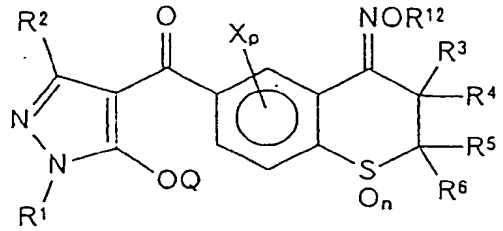
In the general formula (Ia), R^{11} is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_3~C_6 cycloalkyl group, a C_3~C_6 alkenylalkyl group, a C_3~C_6 alkynylalkyl or a C_3~C_6 haloalkenylalkyl group, and preferred is a C_1~C_4 alkyl group. Specific examples of the C_1~C_4 alkyl group, the C_1~C_4 haloalkyl group and the C_3~C_6 cycloalkyl group are as described concerning R^1, R^2 or Y. The C_1~C_4 alkyl group is specifically preferably methyl or ethyl. Specific examples of the C_3~C_6 alkenylalkyl group include $-CH_2-CH=CH_2$, $-CH_2CH=C(CH_3)_2$, $-CH_2C(CH_3)=CH_2$ and $-CH_2-CH=CH-C_2H_5$. Specific examples of the C_3~C_6 alkynylalkyl group include CH2-C≡CH, $-CH_2-C\equiv C-C_2H_5$ and $-CH(CH_3)-C\equiv CH$. The C_3~C_6 haloalkenylalkyl group is a group in which at least one hydrogen atom of the C_3~C_6 alkenylalkyl group is replaced with a halogen atom (e.g., a chlorine atom, a fluorine atom, a bromine atom or an iodine atom), and examples thereof include - $CH_2-CH=CHCl$, $-CH_2-CCl=CH_2$, $-CH_2-CF=CH_2$, - $CH_2CCl=CHCl$, - $CH_2CF=CHF$, and $-CH_2CCl=CH-CH_3$. Preferred embodiments of Z including the above R^{11} are as follows.



The pyrazole derivative in which z is

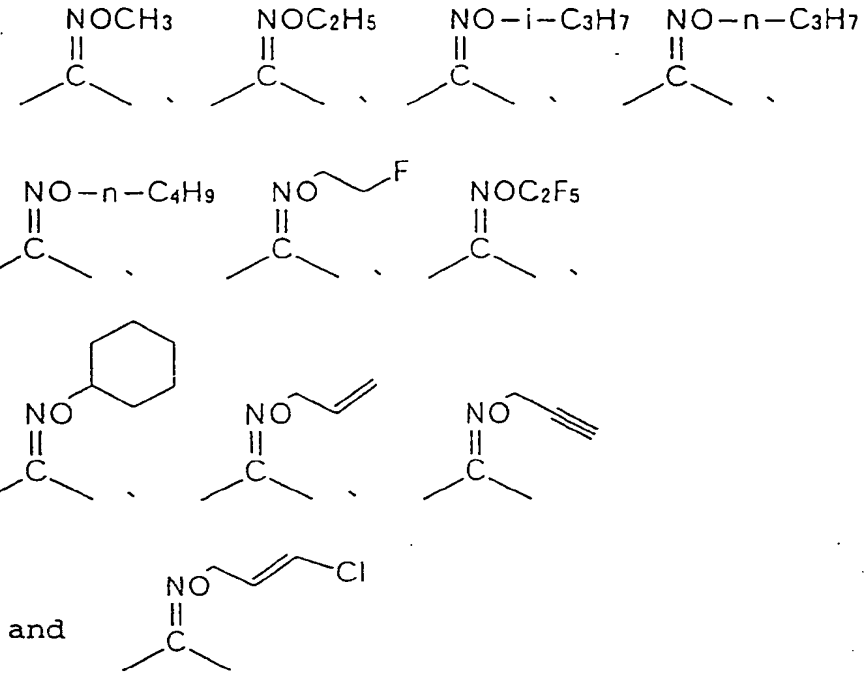


has the following general formula (Ib).

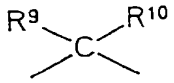


(Ib)

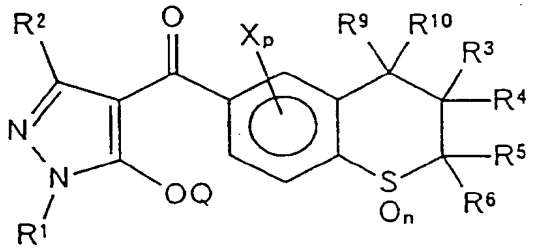
In the general formula (Ib), R^{12} is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_3~C_6 cycloalkyl group, a C_3~C_6 alkenylalkyl group, a C_3~C_6 alkynylalkyl group or a C_3~C_6 haloalkenylalkyl group, and a C_1~C_4 alkyl group is preferred. Specific examples of the C_1~C_4 alkyl group, the C_1~C_4 haloalkyl group, the C_3~C_6 cycloalkyl group, the C_3~C_6 alkenylalkyl group, the C_3~C_6 alkynylalkyl group and the C_3~C_6 haloalkenylalkyl group are as described concerning R^1, R^2, Y or R^{11}. The C_1~C_4 alkyl group is specifically preferably methyl. Preferred embodiments of Z including R^{12} are as follows.



The pyrazole derivative in which Z is

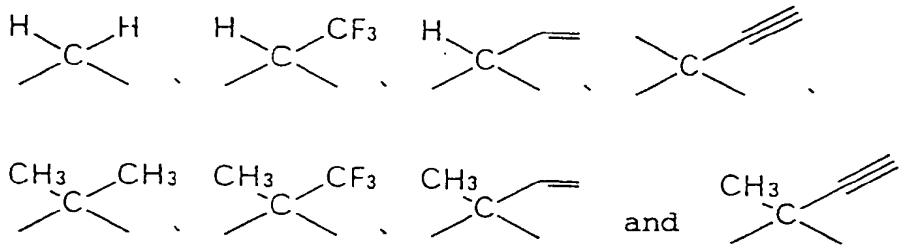


has the following general formula (Ic).

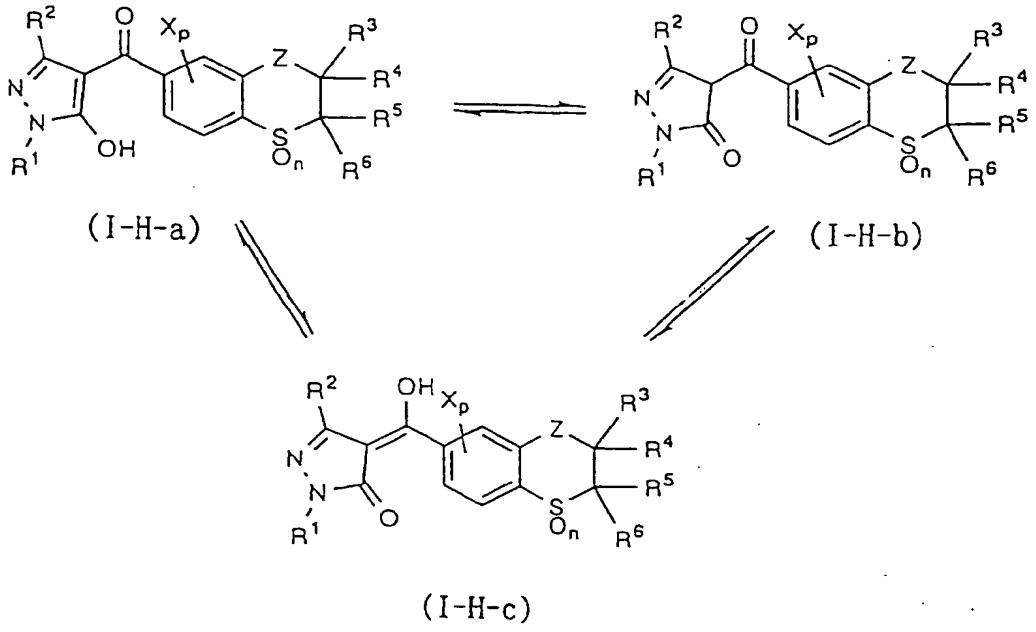


(Ic)

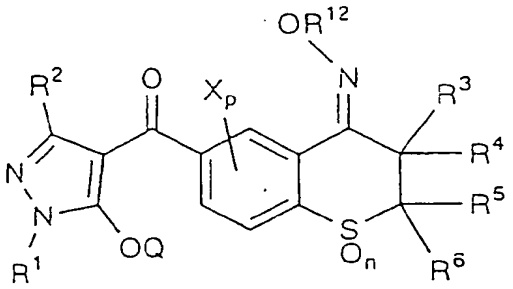
In the general formula (Ic), R^9 is a hydrogen atom, a C_1~C_4 alkyl group or a C_1~C_4 haloalkyl group, and R^{10} is a hydrogen atom, a C_1~C_4 alkyl group, a C_2~C_4 alkenyl group or a C_2~C_4 alkynyl group. Each of these is independently preferably a hydrogen atom or a C_1~C_4 alkyl group. Preferred embodiments of Z including the above R^9 and R^{10} are as follows.



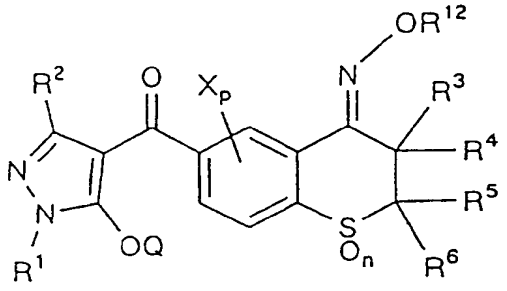
The pyrazole derivative of the general formula (I) in which Q is a hydrogen atom includes the following three tautomers, and all of these are included in the pyrazole derivative of the present invention.



Further, the above pyrazole derivative (Ic) includes the following two geometrical isomers, and both of these are included in the pyrazole derivative of the present invention.



(Ic-α)



(Ic-β)

Some of pyrazole derivatives of the general formula (I) have asymmetric carbon, and include a variety of isomers, and the pyrazole derivative of the present invention includes all of such isomers or mixtures of isomers.

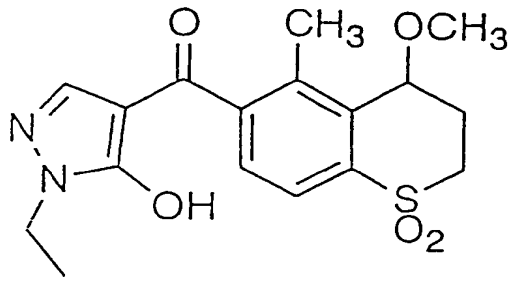
Further, the pyrazole derivative of the general formula (I) in which Q is a hydrogen atom is acidic, and can be easily converted to a salt by treating it with a base. This salt is also included in the pyrazole derivative of the present invention.

As the above base, any known base may be used without any special limitation. Examples thereof include organic bases such as amines and anilines and inorganic bases such as a sodium compound and a potassium compound. The amines include monoalkylamine, dialkylamine and trialkylamine. The alkyl group of each of the alkylamines is generally a C_1~C_4 alkyl group. The anilines include aniline, monoalkylaniline and dialkylaniline. The alkyl group of each of the alkylanilines is generally a C_1~C_4 alkyl group. The sodium compound includes sodium hydroxide and sodium carbonate. The potassium compound includes sodium hydroxide and sodium carbonate.

Preferably, specific examples, chemical names and structural formulae of pyrazole derivatives of the general formula (Ia) are as follows.

Compound(Ia-1)

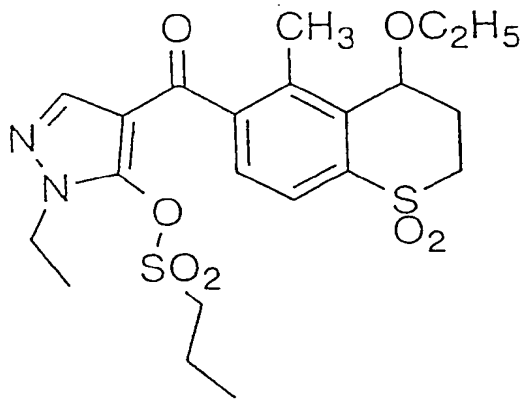
4-methoxy-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{11}=CH_3、Q=H、n=2、p=1)

Compound(Ia-2)

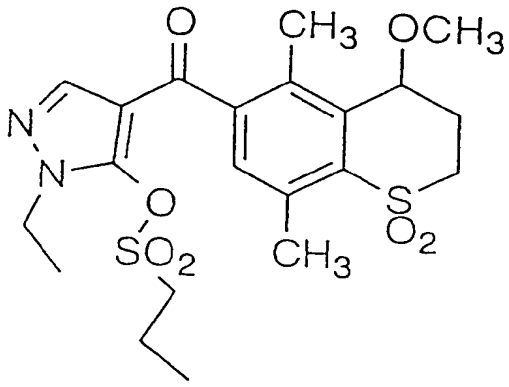
4-ethoxy-5-methyl-6-(1-ethyl-5-n-propanesulfonyloxyprazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{11}=C_2H_5、Q=-SO_2-n-C_3H_7、n=2、p=1)

Compound(Ia-3)

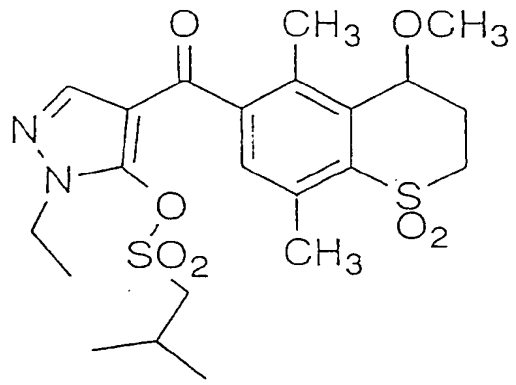
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^{11}=CH_3、Q=-SO_2-n-C_3H_7、n=2、p=2)

Compound(Ia-4)

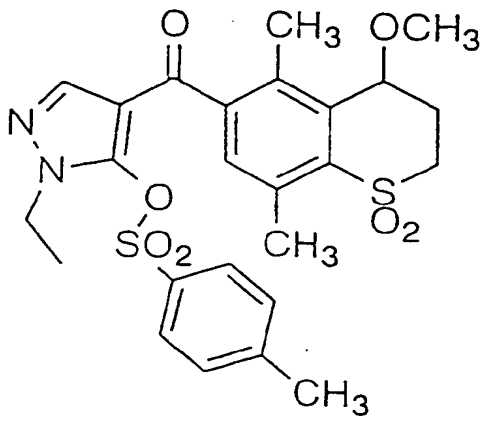
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-i-butanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^{11}=CH_3$、Q=-SO_2-i-C_4H_9、n=2、p=2)

Compound(Ia-5)

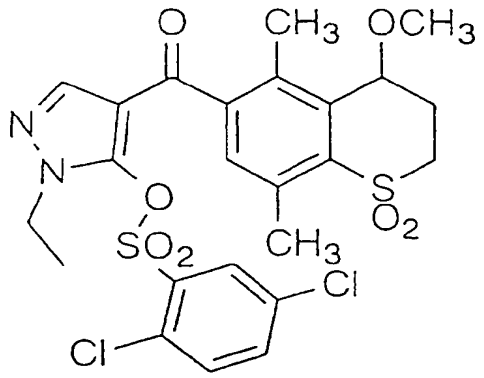
4-methoxy-5,8-dimethl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^{11}=CH_3$、Q=-SO_2-C_6H_4-CH_3、n=2、p=2)

Compound(Ia-6)

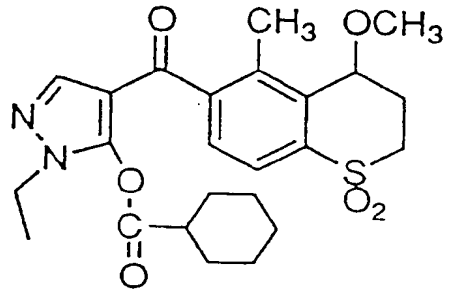
4-methoxy-5,8-dimethyl-6-(1-ethyl-5-(2,5-dichlorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ia)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^{11}=CH_3$、Q=-SO_2-C_6H_3-Cl_2、n=2、p=2)

Compound(Ia-7)

4-methoxy-5-methyl-6-(1-ethyl-5-cyclohexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide

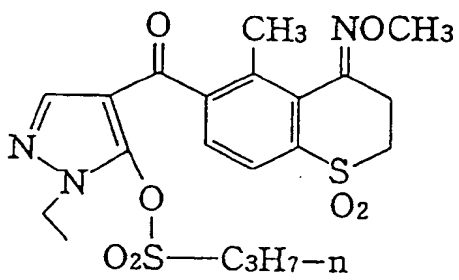


(In the general formula (Ia)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-CH_3、$R^{11}=CH_3$、Q=-CO-cyclo-C_6H_{11}、n=2、p=1)

Preferably, specific examples, chemical names and structural formulae of pyrazole derivative of the general formula (Ib) are as follows.

Compound(Ib-1)

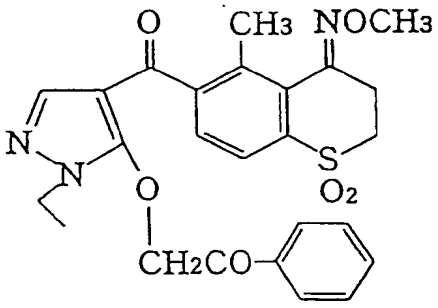
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-SO_2-n-C_3H_7、n=2、p=1)

Compound(Ib-2)

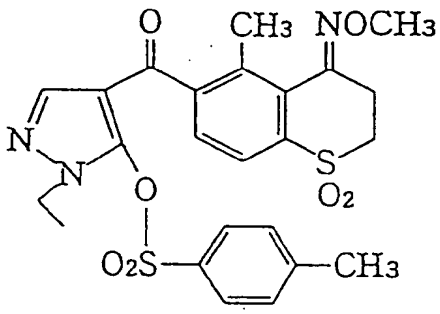
4-methoxyimino-5-methyl-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-CH_2CO-C_6H_5、n=2、p=1)

Compound(Ib-3)

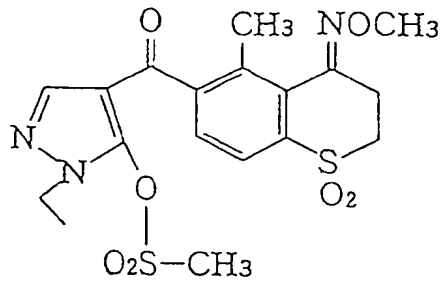
4-methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5-CH_3、$R^{12}=CH_3$、Q=-SO_2-C_6H_4-CH_3、n=2、p=1)

Compound(Ib-4)

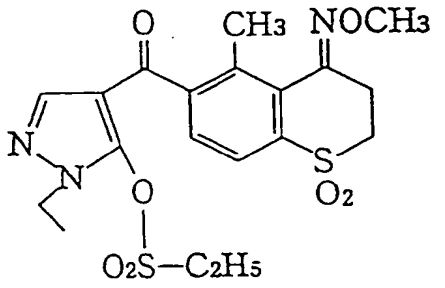
4-methoxyimino-5-methyl-6-(1-ethyl-5-methanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5-CH_3、$R^{12}=CH_3$、Q=SO_2-CH_3、n=2、p=1)

Compound(Ib-5)

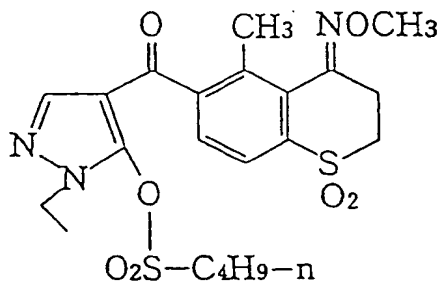
4-methoxyimino-5-methyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5-CH_3、$R^{12}=CH_3$、Q=-SO_2-C_2H_5、n=2、p=1)

Compound(Ib-6)

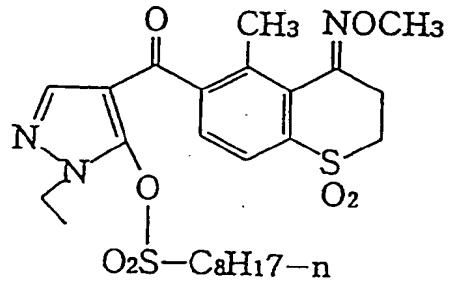
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-butanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-CH_3、$R^{12}=CH_3$、Q=-SO_2-n=C_4H_9、n=2、p=1)

Compound(Ib-7)

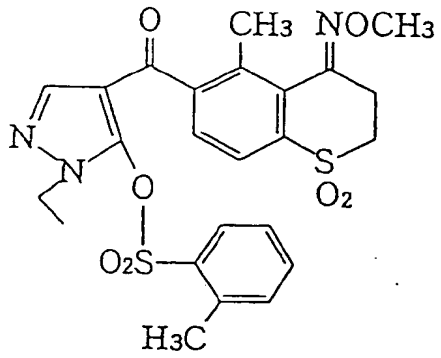
4-methoxyimino-5-methyl-6-(1-ethyl-5-n-octanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-CH_3、$R^{12}=CH_3$、Q=-SO_2-n=C_8H_{17}、n=2、p=1)

Compound(Ib-8)

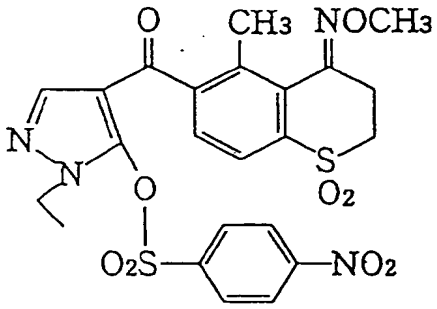
4-methoxyimino-5-methyl-6-(1-ethyl-5-(2-methylphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、 $R^1=C_2H_5$、 $R^2=H$、 $R^3=R^4=R^5=$ $R^6=H$、 X=5-CH_3、 $R^{12}=CH_3$、 Q=-SO_2-C_6H_4-CH_3、 n=2、 p=1)

Compound(Ib-9)

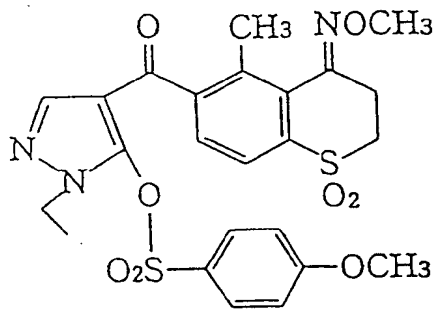
4-methoxyimino-5-methyl-6-(1-ethyl-5-(2-nitrophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、 $R^1=C_2H_5$、 $R^2=H$、 $R^3=R^4=R^5=$ $R^6=H$、 X=5-CH_3、 $R^{12}=CH_3$、 Q=-SO_2-C_6H_4-NO_2、 n=2、 p=1)

Compound (Ib-10)

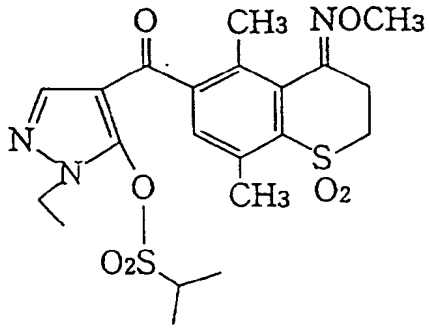
4-methoxyimino-5-methyl-6-(1-ethyl-5-(4-methoxyphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-SO_2-C_6H_4-OCH_3、n=2、p=1)

Compound(Ib-11)

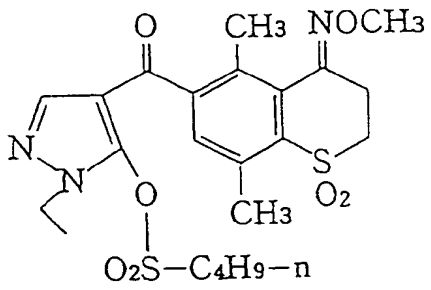
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-i-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^{12}=CH_3、Q=-SO_2-i-C_3H_7、n=2、p=2)

Compound(Ib-12)

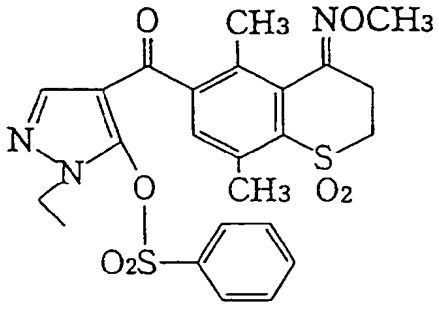
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-n-butanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^{12}=CH_3、Q=-SO_2-n-C_4H_9、n=2、p=2)

Compound(Ib-13)

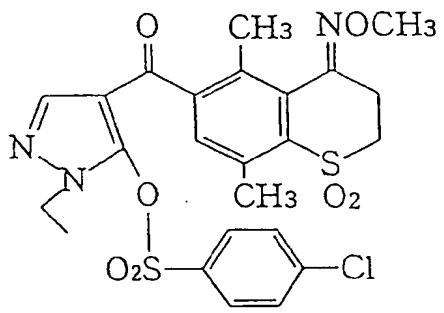
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-phenylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^{12}=CH_3、Q=-SO_2-C_6H_5、n=2、p=2)

Compound(Ib-14)

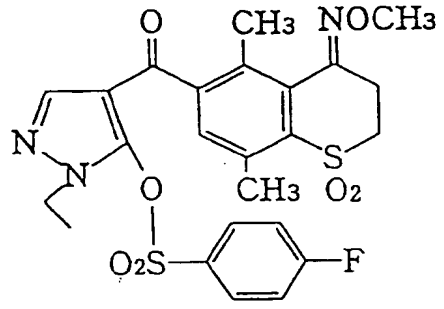
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-(4-chlorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、Q=-SO_2-C_6H_4-Cl、n=2、p=2)

Compound(Ib-15)

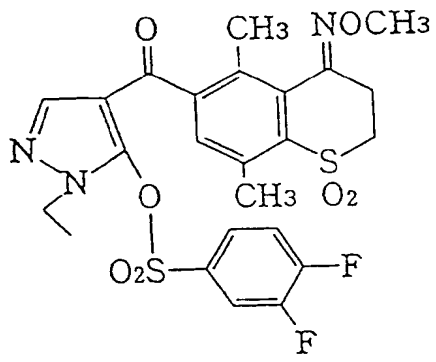
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-(4-fluorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、Q=-SO_2-C_6H_4-F、n=2、p=2)

Compound(Ib-16)

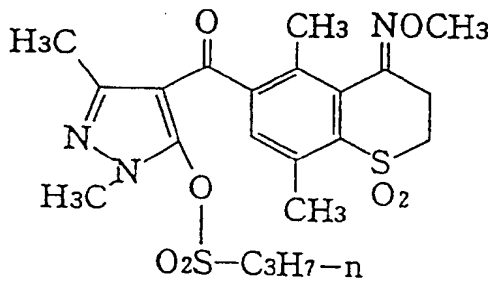
5,8-dimethyl-4-methoxyimino-(1-ethyl-5-(3,4-difluorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、Q=-SO_2-C_6H_3-F_2、n=2、p=2)

Compound(Ib-17)

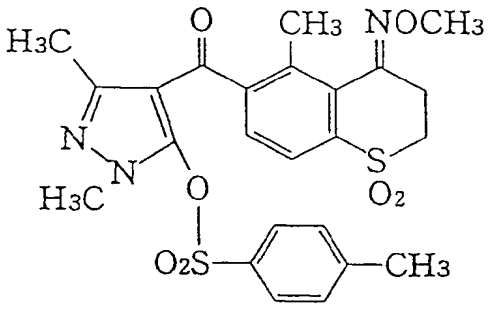
5,8-dimethyl-4-methoxyimino-(1,3-dimethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=CH_3$、$R^2=CH_3$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、Q=-SO_2-n-C_3H_7、n=2、p=2)

Compound(Ib-18)

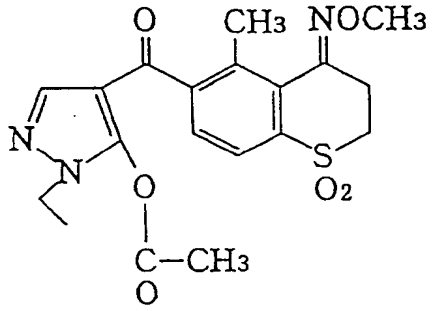
4-methoxyimino-5-methyl-(1,3-dimethyl-5-p-toluensulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=CH_3、R^2=CH_3、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-SO_2-C_6H_4-CH_3、n=2、p=1)

Compound(Ib-19)

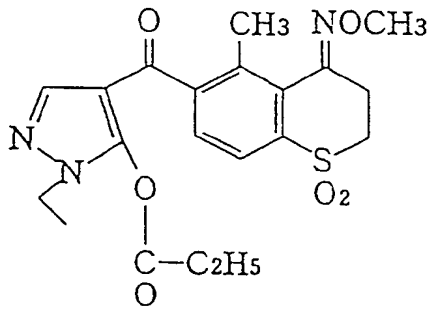
4-methoxyimino-5-methyl-6-(1-ethyl-5-acetyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-CO-CH_3、n=2、p=1)

Compound(Ib-20)

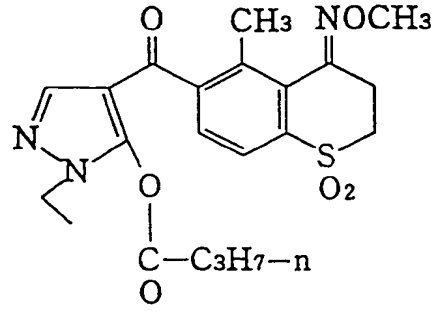
4-methoxyimino-5-metyl-6-(1-ethyl-5-ethanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-CH_3、$R^{12}=CH_3$、Q=-CO-C_2H_5、n=2、p=1)

Compound(Ib-21)

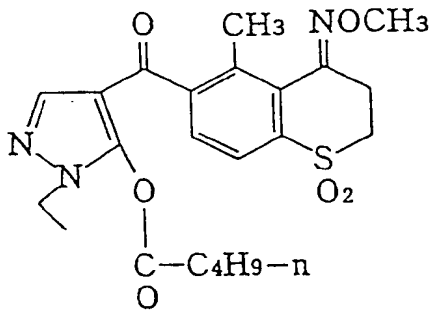
4-methoxyimino-5-metyl-6-(1-ethyl-5-n-propanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-CH_3、$R^{12}=CH_3$、Q=-CO-n-C_3H_7、n=2、p=1)

Compound(Ib-22)

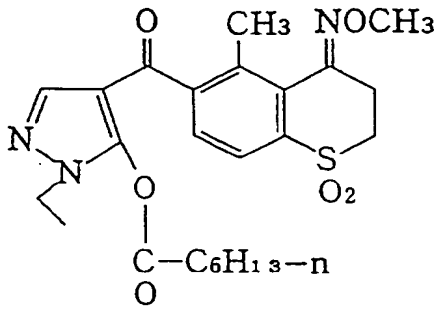
4-methoxyimino-5-metyl-6-(1-ethyl-5-n-butanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-CO-n-C_4H_9、n=2、p=1)

Compound(Ib-23)

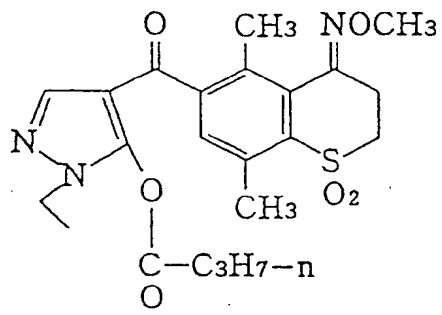
4-methoxyimino-5-metyl-6-(1-ethyl-5-n-hexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^{12}=CH_3、Q=-CO-n-C_6H_{13}、n=2、p=1)

Compound(Ib-24)

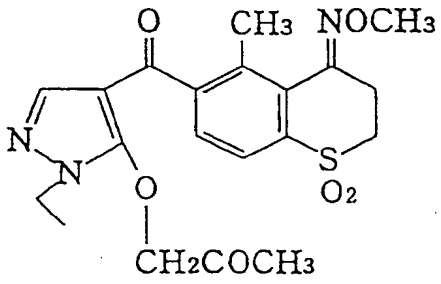
5,8-dimethyl-4-methoxyimino-6-(1-ethyl-5-n-propanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、Q=-CO-n-C_3H_7、n=2、p=2)

Compound(Ib-25)

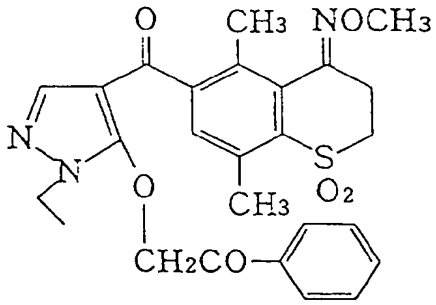
4-methoxyimino-5-methyl-6-(1-ethyl-5-acetylmethyleneoxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-CH_3、$R^{12}=CH_3$、Q=-CH_2COCH_3、n=2、p=1)

Compound(Ib-26)

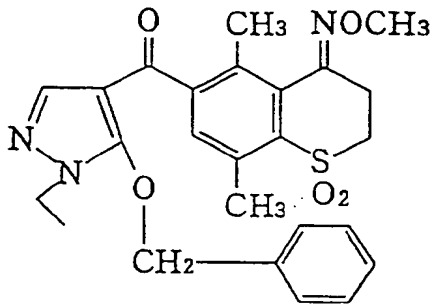
5,8-dimethyl-4-methoxyimino-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=$ $R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、$Q=CH_2CO\text{-}C_6H_5$、n=2、p=2)

Compound(Ib-27)

5,8-dimethyl-4-methoxyimino-6-(1-ethyl-5-benzyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide

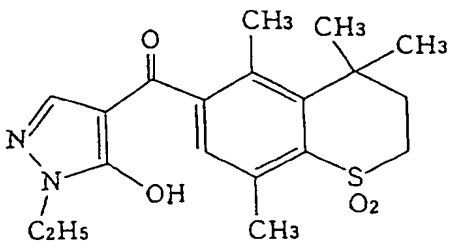


(In the general formula (Ib)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=$ $R^6=H$、X=5, 8-CH_3、$R^{12}=CH_3$、$Q=CH_2\text{-}C_6H_5$、n=2、p=2)

Preferably, specific examples, chemical names and structural formulae of pyrazole derivative of the general formula (Ic) are as follows.

Compound(Ic-1)

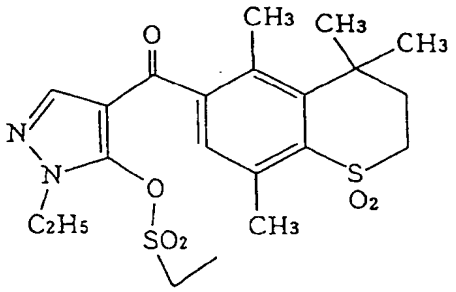
4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxylpyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=H、n=2、p=2)

Compound(Ic-2)

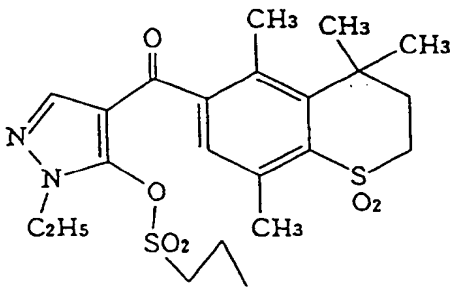
4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-C_2H_5、n=2、p=2)

Compound(Ic-3)

4,4,5,8-tetramethyl-6-(1-ethyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide

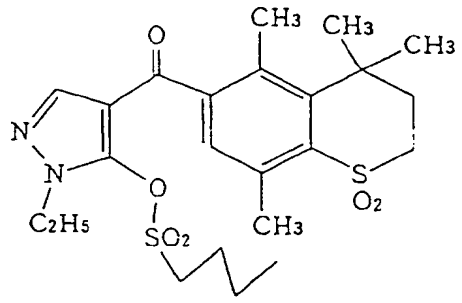


(In the general formula (Ic)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-n-C_3H_7、

n=2、p=2)

Compound(Ic-4)

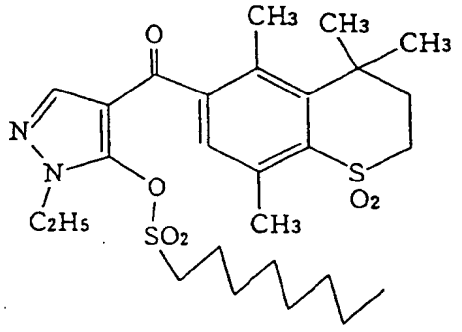
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-butanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-n-C_4H_9、n=2、p=2)

Compound(Ic-5)

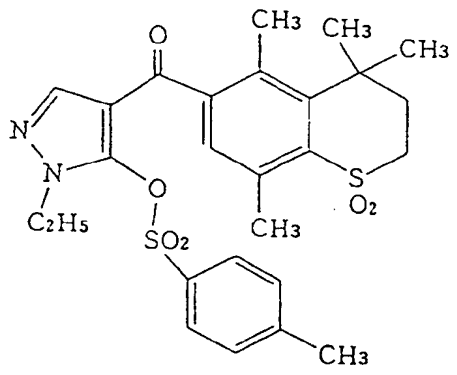
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-octanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-C_8H_{17}、n=2、p=2)

Compound(Ic-6)

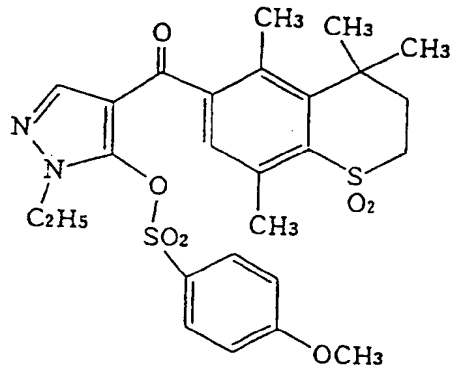
4,4,5,8-tetramethyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、 $R^1=C_2H_5$、 $R^2=H$、 $R^3=R^4=R^5=R^6=H$、 X=5, 8-CH_3、 $R^9=R^{10}=CH_3$、 Q=-SO_2-C_6H_4-CH_3、 n=2、 p=2)

Compound(Ic-7)

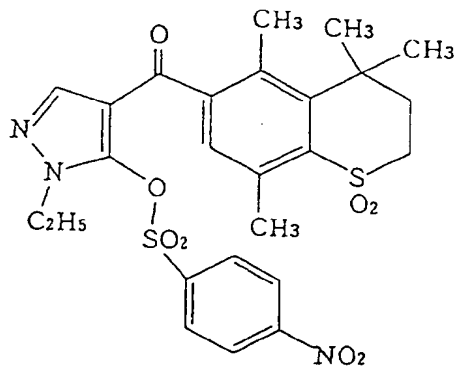
4,4,5,8-tetramethyl-6-(1-ethyl-5-(4-methoxyphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、 $R^1=C_2H_5$、 $R^2=H$、 $R^3=R^4=R^5=R^6=H$、 X=5, 8-CH_3、 $R^9=R^{10}=CH_3$、 Q=-SO_2-C_6H_4-OCH_3、 n=2、 p=2)

Compound(Ic-8)

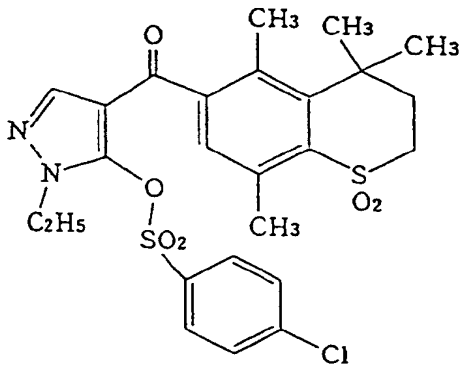
4,4,5,8-tetramethyl-6-(1-ethyl-5-(4-nitrophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-C_6H_4-NO_2、n=2、p=2)

Compound(Ic-9)

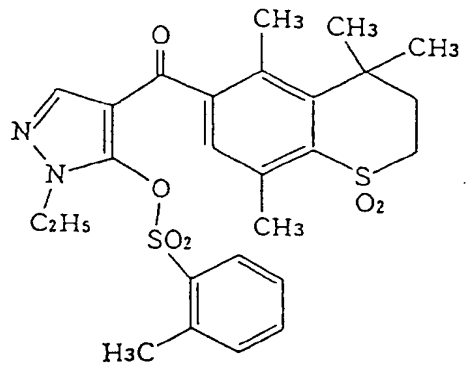
4,4,5,8-tetramethyl-6-(1-ethyl-5-p-chlorophenylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-C_6H_4-Cl、n=2、p=2)

Compound(Ic-10)

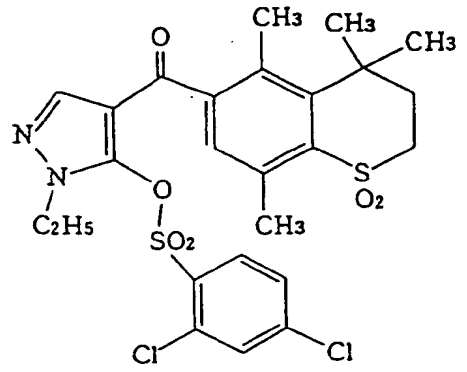
4,4,5,8-tetramethyl-6-(1-ethyl-5-(2-methylphenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-C_6H_4-CH_3、n=2、p=2)

Compound(Ic-11)

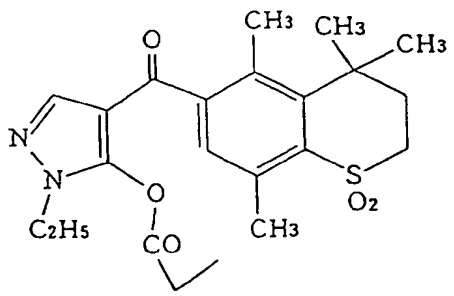
4,4,5,8-tetramethyl-6-(1-ethyl-5-(2,4-dichlorophenyl)sulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-SO_2-C_6H_3-Cl、n=2、p=2)

Compound(Ic-12)

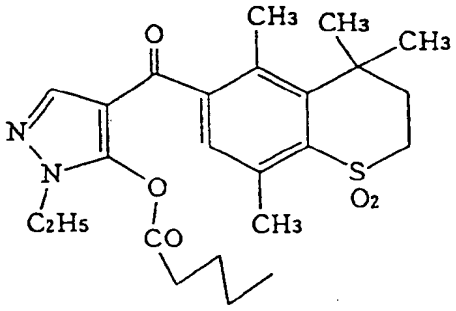
4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=$ $R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-CO-C_2H_5、n=2、p=2)

Compound(Ic-13)

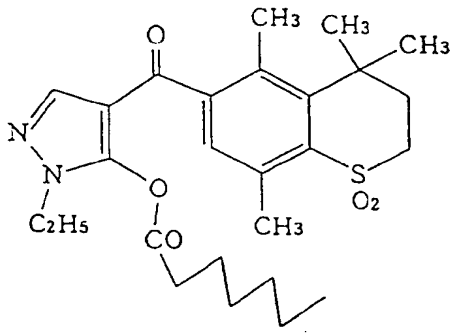
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-butanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=$ $R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-CO-n-C_4H_9、n=2、p=2)

Compound(Ic-14)

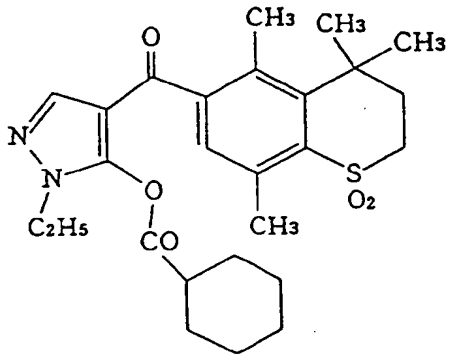
4,4,5,8-tetramethyl-6-(1-ethyl-5-n-hexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-CO-n-C_6H_{13}、n=2、p=2)

Compound(Ic-15)

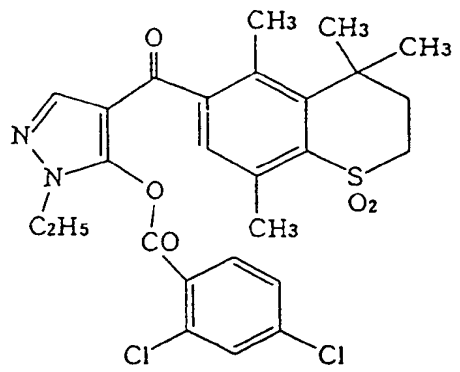
4,4,5,8-tetramethyl-6-(1-ethyl-5-cyclohexanecarbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-CO-cyclo-C_6H_{11}、n=2、p=2)

Compound(Ic-16)

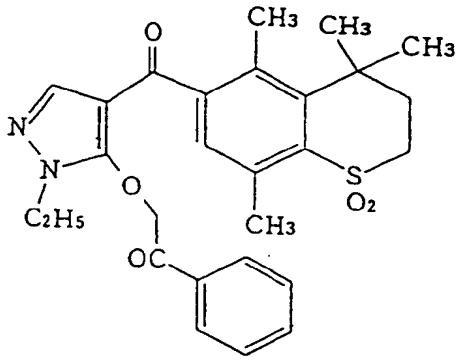
4,4,5,8-tetramethyl-6-(1-ethyl-5-(2,4-dichlorophenyl)carbonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-CO-C_6H_4-Cl_2、n=2、p=2)

Compound(Ic-17)

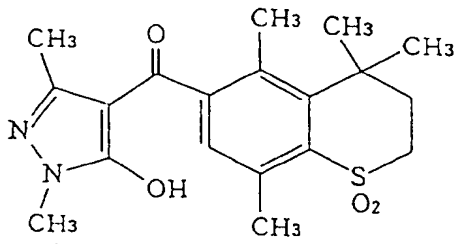
4,4,5,8-tetramethyl-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、R^2=H、$R^3=R^4=R^5=R^6$=H、X=5, 8-CH_3、$R^9=R^{10}=CH_3$、Q=-CO-C_6H_5、n=2、p=2)

Compound(Ic-18)

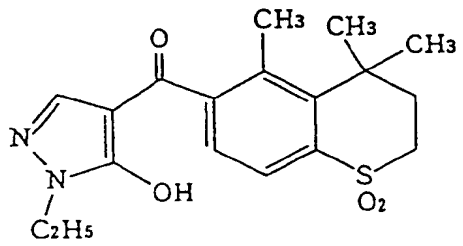
4,4,5,8-tetramethyl-(1,3-dimethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、R^1=CH_3、R^2=CH_3、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^9=R^{10}=CH_3、Q=H、n=2、p=2)

Compound(Ic-19)

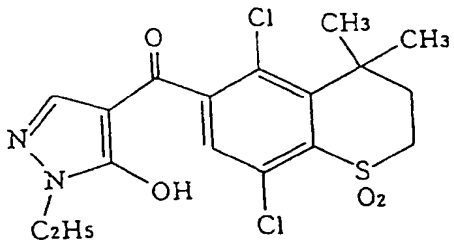
4,4,5-trimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5-CH_3、R^9=R^{10}=CH_3、Q=H、n=2、p=1)

Compound(Ic-20)

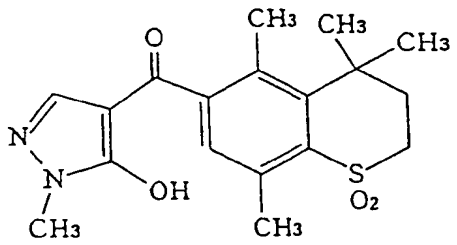
5,8-dichloro-4,4-dimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、R^1=C_2H_5、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-Cl、R^9=R^{10}=CH_3、Q=H、n=2、p=2)

Compound(Ic-21)

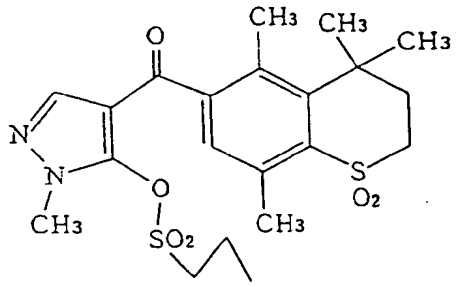
4,4,5,8-tetramethyl-6-(1-methyl-5-hydroxypyrazol-4-yl)carbonylthiocnroman-1,1-dioxide



(In the general formula (Ic)、R^1=CH_3、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^9=R^{10}=CH_3、Q=H、n=2、p=2)

Compound(Ic-22)

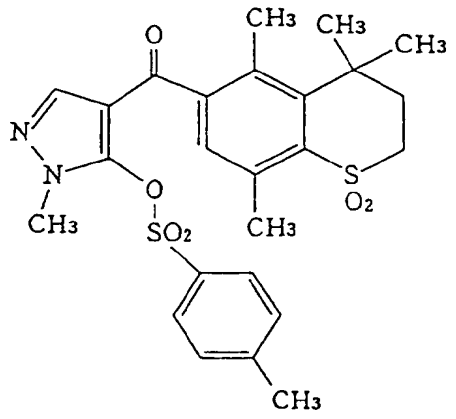
4,4,5,8-tetramethyl-6-(1-methyl-5-n-propanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、R^1=CH_3、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^9=R^{10}=CH_3、Q=-SO_2-n-C_3H_7、n=2、p=2)

Compound(Ic-23)

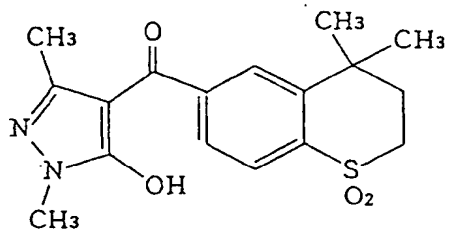
4,4,5,8-tetramethyl-6-(1-methyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide



(in the general formula (Ic)、R^1=CH_3、R^2=H、R^3=R^4=R^5= R^6=H、X=5, 8-CH_3、R^9=R^{10}=CH_3、Q=-SO_2-C_6H_4-CH_3、n=2、p=2)

Compound(Ic-24)

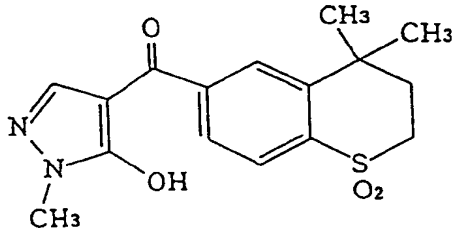
4,4-dimethyl-(1,3-dimethyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、R^1=CH_3、R^2=CH_3、R^3=R^4=R^5= R^6=H、R^9=R^{10}=CH_3、Q=H、n=2、p=0)

Compound(Ic-25)

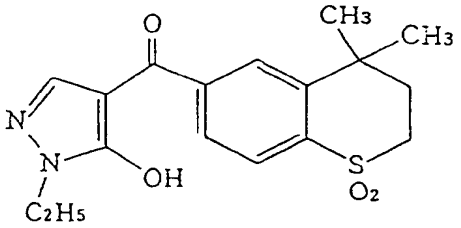
4,4-dimethyl-(1-methyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=CH_3$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、$R^9=R^{10}=CH_3$、Q=H、n=2、p=0)

Compound(Ic-26)

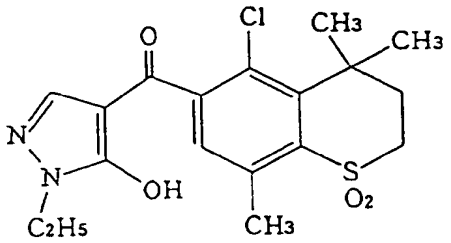
4,4-dimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、$R^9=R^{10}=CH_3$、Q=H、n=2、p=0)

Compound(Ic-27)

5-chloro-4,4,8-trimethyl-(1-ethyl-5-hydroxypyrazol-4-yl)-carbonylthiochroman-1,1-dioxide



(In the general formula (Ic)、$R^1=C_2H_5$、$R^2=H$、$R^3=R^4=R^5=R^6=H$、X=5-Cl、8-CH_3、$R^9=R^{10}=CH_3$、Q=H、n=2、p=2)

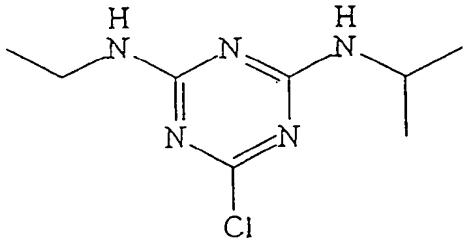
In the herbicide composition of the present invention, the herbicide compound which is the second active ingredient can exhibit a synergistic effect when used with the above mentioned pyrazole derivative (I). Examples of this herbicide compound includes germination inhibitor such as chloroacetamide herbicide or dinitroaniline herbicide; plant hormone such as benzoic acid herbicide, pyridine carboxylic acid herbicide or phenoxy herbicide; ALS (Acetolactate Synthetase) inhibitor such as surfonylurea herbicide or imidazolinone herbicide; photosynthesis inhibitor such as triazine herbicide or urea herbicide; and other herbicide such as diazine herbicide, benzonitrile herbicide or pyridazine herbicide.

In the herbicide composition of the present invention, among the above mentioned herbicide compound, at least one member selected from the following groups is used as an active ingredient with the pyrazole derivative (I).

Chloroacetamide herbicide;
Imidazolinone herbicide;
Triazine herbicide such as Compound (B-1) ~ (B-3);

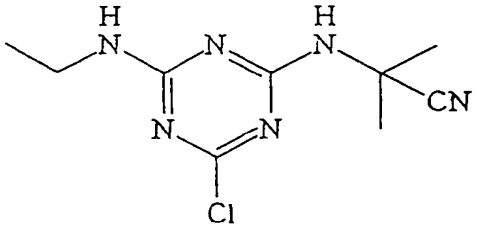
Compound(B-1)

Common name:atrazine
Chemical Name:
6-chloro-N^2-ethyl-N^4-isopropil-1,3,5-triazine-2,4-diamine



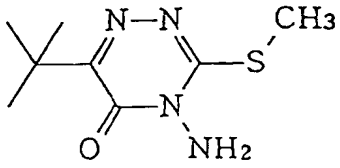
Compound(B-2)

Common name:cyanazine
Chemical Name:
2-(4-chloro-6-ethylamino-1,3,5-triazin-2-ylamino)-2-methylpropionitrile



Compound(B-3)

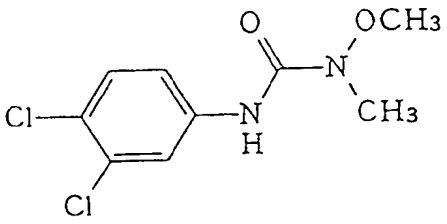
Common name:metribuzin
Chemical Name:
4-amino-6-tert-buthyl-4,5-dihydro-3-methylthio-1,2,4-triazin-5-one



Urea herbicide such as Compounds (B-4) and (B-5) ;

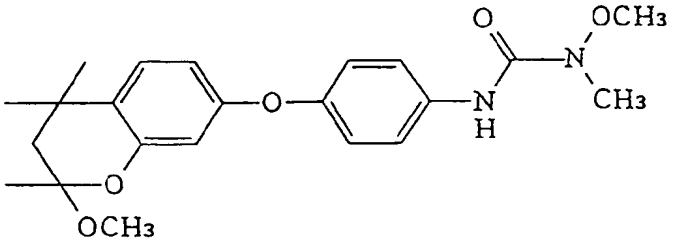
Compound(B-4)

Common name:linuron
Chemical Name:
3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea



Compound(B-5)

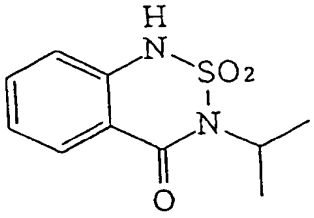
Common name:metbenzurone
Chemical Name:
(±)-1-methoxy-3-[4-(2-methoxy-2,4,4-trimethylchroman-7-yloxy)phenyl]-1-methylurea



Diazine herbicide such as Compound (B-6) ;

Compound(B-6)

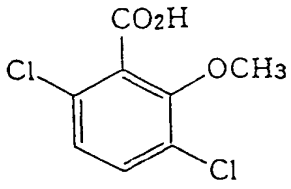
Common name:bentazone
Chemical Name:
3-isopropyl-1H-2,1,3-benzothiadiazin-4(3H)-one-2,2-dioxide



Benzoic acid herbicide such as Compound (B-7);

Compound(B-7)

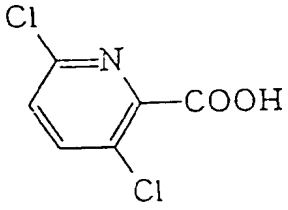
Common name:dicamba
Chemical Name:
3,6-dichloro-2-methoxybenzoic acid



Pyridine carboxylic acid herbicide such as Compound (B-8) ;

Compound(B-8)

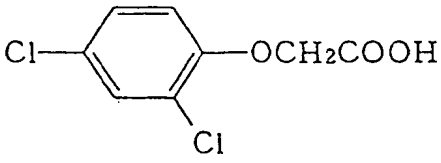
Common name:chlopyralid
Chemical Name:
3,6-dichloropyridine-2-carboxylic acid



Phenoxy herbicide such as Compound (B-9) ;

Compound(B-9)

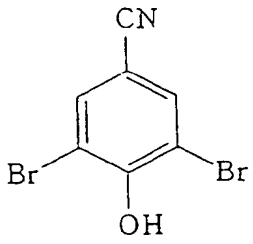
Common name:2,4-D
Chemical Name:
2-(2,4-dichlorophenoxy)acetic acid



Benzonitrile herbicide such as Compound (B-10) ;

Compound(B-10)

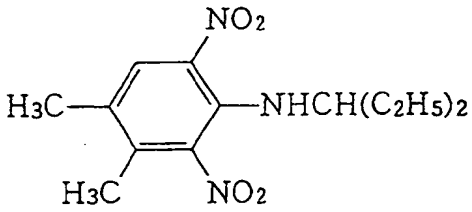
Common name:bromoxynil
Chemical Name:
3,5-dibromo-4-hydroxybenzonitrile



Dinitroaniline herbicide such as Compound (B-13) ;

Compound(B-13)

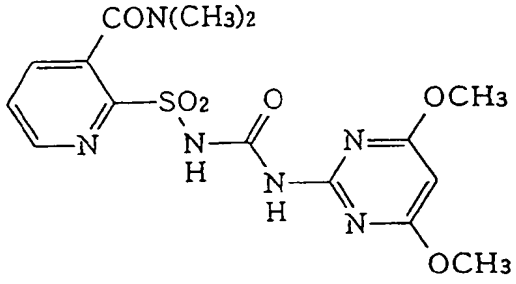
Common name:pendimethalin
Chemical Name:
N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine



Sulfonyl urea herbicide such as Compound (B-14), (B-15) and (B-17) ;

Compound(B-14)

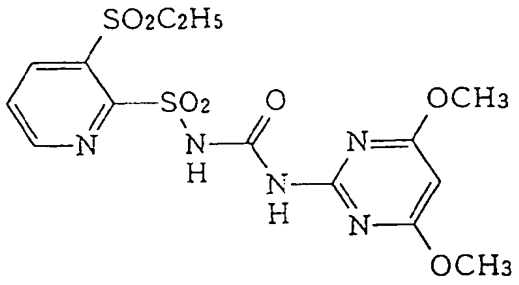
Common name:nicosulfuron
Chemical Name:
2-(4,6-dimethoxypyrimidin-2-ylcarbamoylsulfamoyl)-N,N-dimethylnicotineamide



Compound(B-15)

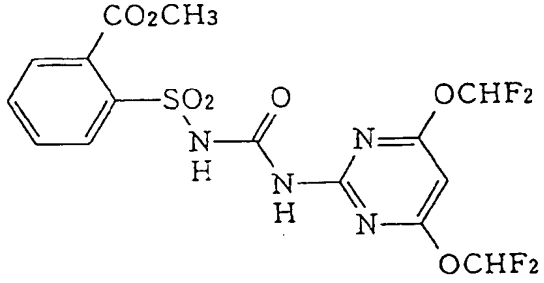
Common name:rimsulfuron
Chemical Name:

1-(4,6-dimethoxypyrimidin-2-yl-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea



Compound(B-17)

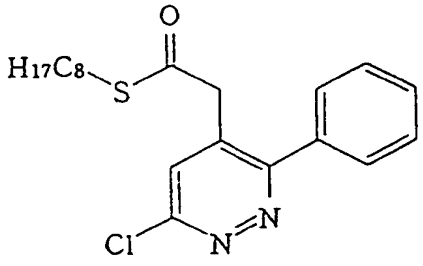
Common name:primisulfuron
Chemical Name:
Methyl 2-[4,6-bis(difluoromethoxy)pyrimidin-2-yl-carbamoylsulfamoyl]benzoic acid



Pyridazin herbicide such as Compound (B-20) ;

Compound(B-20)

Common name:pyridate
Chemical Name:
6-chloro-3-phenylpyridazin-4-yl-S-octylthiocarbonate

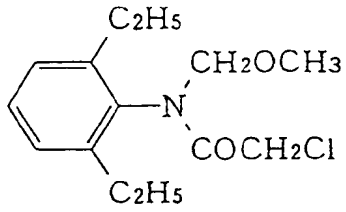


Among the herbicide compounds, the following Compounds (B-11), (B-12), (B-18) and (B-19) are exemplified as

the chloroacetamide herbicide.

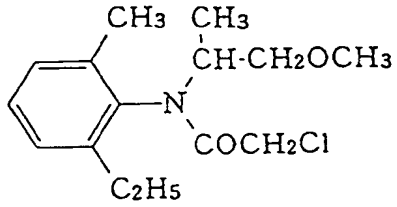
Compound(B-11)

Common name:alachlor
Chemical Name:
2-chloro-2',6'-diethyl-N-methoxymethylacetanilide



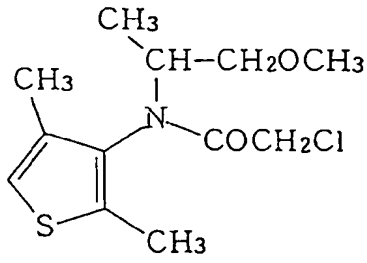
Compound(B-12)

Common name:metolachlor
Chemical Name:
2-chloro-N-(2-ethyl-6-methylphenyl)-N-(2-methoxy-1-methylethyl)acetamide



Compound(B-18)

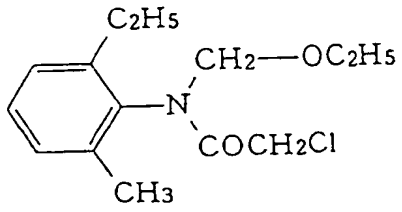
Common name:dimethenamid
Chemical Name:
(1RS,aRS)-2-chloro-N-(2,4-dimethyl-3-thienyl)-N-(2-methoxy-1-methylethyl)acetamide



Compound(B-19)

Common name:acetochlor
Chemical Name:

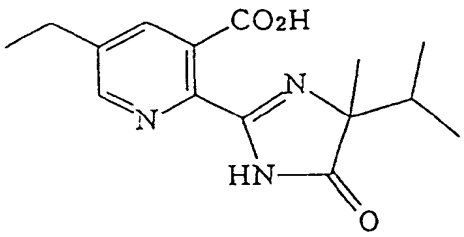
2-chloro-2'-ethyl-6'-methyl-N-ethoxymethylacetanilide



Among the herbicide compounds, the following Compounds (B-16) and (B-21) are exemplified as the imidazolinone herbicide.

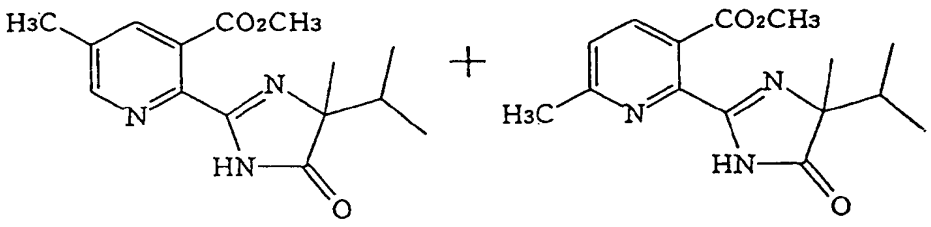
Compound(B-16)

Common name:imazethapyr
Chemical Name:
5-ethyl-2-(4-isopropil-4-methyl-5-oxo-2-imidazolin-2-yl)nicotinic acid



Compound(B-21)

Common name:imazamethabenz-methyl
Chemical Name:
Compoud of a Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin -2-yl)-m-toluic acid and a Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-p-toluic acid



(Imazamethabenz-methyl which is a mixture of the two isomers is commercially available under a trade name of Assert.)

The herbicide composition of the present invention contains, as active ingredients, at least one of the above pyrazole derivatives (I) and at least one selected from a group of the above herbicide compounds. The mixing ratio of these is not specially limited, and a synergistic effect can be obtained in a broad range of the mixing ratio, while the pyrazole derivative (I) and the compounds (B-1)~(B-2) are generally preferably mixed in the following mixing ratios (weight

ratios).

pyrazole derivative(I):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(I):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(I):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(I):compound(B-4:linuron)=2:1~1:50
pyrazole derivative(I):compound(B-5:metbenzurone)=1:2~ 1:100
pyrazole derivative(I):compound(B-6:bentazone)=6:1~1:100
pyrazole derivative(I):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(I):compound(B-8:chlopyralid)=4:3~1:12
pyrazole derivative(I):compound(B-9:2,4-D)=2:1~1:5
pyrazole derivative(I):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(I):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(I):compound(B-12:metolachlor)=2:1~1:25
pyrazole derivative(I):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(I):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(I):compound(B-15:rimsulfuron)=1:3~40:1
pyrazole derivative(I):compound(B-16:imazethapyr)=1:6~40:1
pyrazole derivative(I):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(I):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(I):compound(B-19:acetochlor)=2:1~1:50
pyrazole derivative(I):compound(B-20:pyridate)=3:2~1:50
pyrazole derivative(I):compound(B-21:imazamethabenzmethyl)=1:6~40:1

The above pyrazole derivatives (Ia), (Ib) and (Ic) and the compounds (B-1)~(B-20) are preferably mixed in the following mixing ratios, respectively.

pyrazole derivative(Ia):compound(B-1:atrazine)=1:1~1:50
pyrazole derivative(Ia):compound(B-2:cyanazine)=2:1~1:25
pyrazole derivative(Ia):compound(B-3:metribuzin)=3:1~1:12
pyrazole derivative(Ia):compound(B-4:linuron)=2:1~1:25
pyrazole derivative(Ia):compound(B-6:bentazone)=1:2~1:100
pyrazole derivative(Ia):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(Ia):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(Ia):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(Ia):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(Ia):compound(B-12:metolachlor)=2:1~ 1:25
pyrazole derivative(Ia):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(Ia):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(Ia):compound(B-15:rimsulfuron)=1:3~ 40:1
pyrazole derivative(Ia):compound(B-16:imazethapyr)=1:6~ 40:1
pyrazole derivative(Ia):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(Ia):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(Ia):compound(B-19:acetochlor)=2:1~1:50
pyrazole derivative(Ia):compound(B-20:pyridate)=3:2~1:50
pyrazole derivative(Ib):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(Ib):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(Ib):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(Ib):compound(B-4:linuron)=2:1~1:50
pyrazole derivative(Ib):compound(B-5:metbenzurone)=1:2~ 1:100
pyrazole derivative(Ib):compound(B-6:bentazone)=6:1~1:3
pyrazole derivative(Ib):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(Ib):compound(B-8:chlopyralid)=4:3~1:12
pyrazole derivative(Ib):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(Ib):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(Ib):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(Ib):compound(B-12:metolachlor)=2:1~ 1:25
pyrazole derivative(Ib):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(Ib):compound(B-14:nicosulfuron)=1:3~ 40:1

pyrazole derivative(Ib):compound(B-15:rimsulfuron)=1:3~ 40:1
pyrazole derivative(Ib):compound(B-16:imazethapyr)=1:6~ 40:1
pyrazole derivative(Ib):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(Ib):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(Ib):compound(B-19:acetochlor)=2:1~1:50
pyrazole derivative(Ic):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(Ic):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(Ic):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(Ic):compound(B-4:linuron)=2:1~1:50
pyrazole derivative(Ic):compound(B-6:bentazone)=1:2~1:100
pyrazole derivative(Ic):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(Ic):compound(B-8:chlopyralid)=4:3~1:12
pyrazole derivative(Ic):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(Ic):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(Ic):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(Ic):compound(B-12:metolachlor)=2:1~ 1:25
pyrazole derivative(Ic):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(Ic):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(Ic):compound(B-15:rimsulfuron)=1:3~ 40:1
pyrazole derivative(Ic):compound(B-16:imazethapyr)=1:6~ 40:1
pyrazole derivative(Ic):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(Ic):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(Ic):compound(B-19:acetochlor)=2:1~1:50

The process for the production of the herbicide composition of the present invention will be explained hereinafter.

The herbicide composition of the present invention is obtained by mixing the pyrazole derivative of the above general formula (I) and at least one compound selected from a group of the above herbicide compounds with a liquid carrier such as a solvent or a solid carrier such as a mineral powder and forming the mixture into a preparation in the form of a wettable powder, an emulsifiable concentrate, a dust, granules, a flowable preparation or a solution. The preparation can be formed by adding surfactants such as an emulsifier, a dispersing agent, a spreading agent, a systemic agent and a stabilizer and other adjuvants as required.

When the herbicide composition of the present invention is used as a wettable powder, generally, a composition is prepared by 10 to 55 % by weight of the pyrazole derivative (I) and at least one selected from a group of the above herbicide compounds as active ingredients, 40 to 88 % by weight of a solid carrier and 2 to 5% by weight of a surfactant, and can be used as such. Further, when it is used in the forms of an emulsifiable concentrate and a flowable preparation, generally, it can be prepared by 5 to 50 % by weight of the pyrazole derivative (I) and at least one compound selected from a group of the above herbicide compounds as active ingredients, 35 to 90 % by weight of a solvent and 5 to 15 % by weight of a surfactant and other adjuvant.

On the other hand, when herbicide composition of the present invention is used in the form of a dust, generally, it can be prepared by mixing 1 to 15 % by weight of the pyrazole derivative (I) and at least one compound selected from a group of the above herbicide compounds as active ingredients and 85 to 99 % by weight of a solid carrier. Further, when it is used in the form of granules, it can be prepared by mixing 0.1 to 15 % by weight of the pyrazole derivative (I) and at least one compound selected from a group of the above herbicide compounds as active ingredients, 80 to 97.9 % by weight of a solid carrier and 2 to 5 % by weight of a surfactant. The above solid carrier can be selected from mineral fine powders, and the mineral fine powders include oxides such as diatomaceous earth and slaked lime, phosphates such as apatite, sulfates such as gypsum, and silicates such as talc, pyroferrite, clay, kaolin, bentonite, acid clay, white carbon, powdered quartz and powdered silica.

The liquid carrier includes paraffin- or naphthene-based hydrocarbons such as kerosene, mineral oil and spindle oil, aromatic hydrocarbons such as benzene, toluene and xylene, chlorinated hydrocarbons such as o-chlorotoluene, trichloromethane and trichloroethylene, alcohols such as cyclohexanol, amyl alcohol and ethylene glycol, alcoholethers such as ethylene glycol monomethyl ether and ethylene glycol monoethyl ether, ketones such as isophorone, cyclohexanone and cyclohexenyl-cyclohexanone, ethers such as butyl cellosolve, dimethyl ether and methyl ethyl ether, esters such as isopropyl acetate, benzyl acetate and methyl phthalate, amides such as dimethylformamide, nitriles such as acetonitrile and propionitrile, sulfoxides such as dimethylsulfoxde, organic solvents such as mixtures of these, and water.

Further, the surfactant can be selected from anionic ones (alkylbenzenesulfonate, alkylsulfonate and lauric amide sulfonate), nonionic ones (polyoxyethylene octyl ether, polyethylene glycol laurate and sorbitan alkyl ester), cationic ones (dimethyllaurylbenzylammonium chloride, laurylamine and stearyltrimethylammonium chloride) and amphoteric ones (amino acid and betaine).

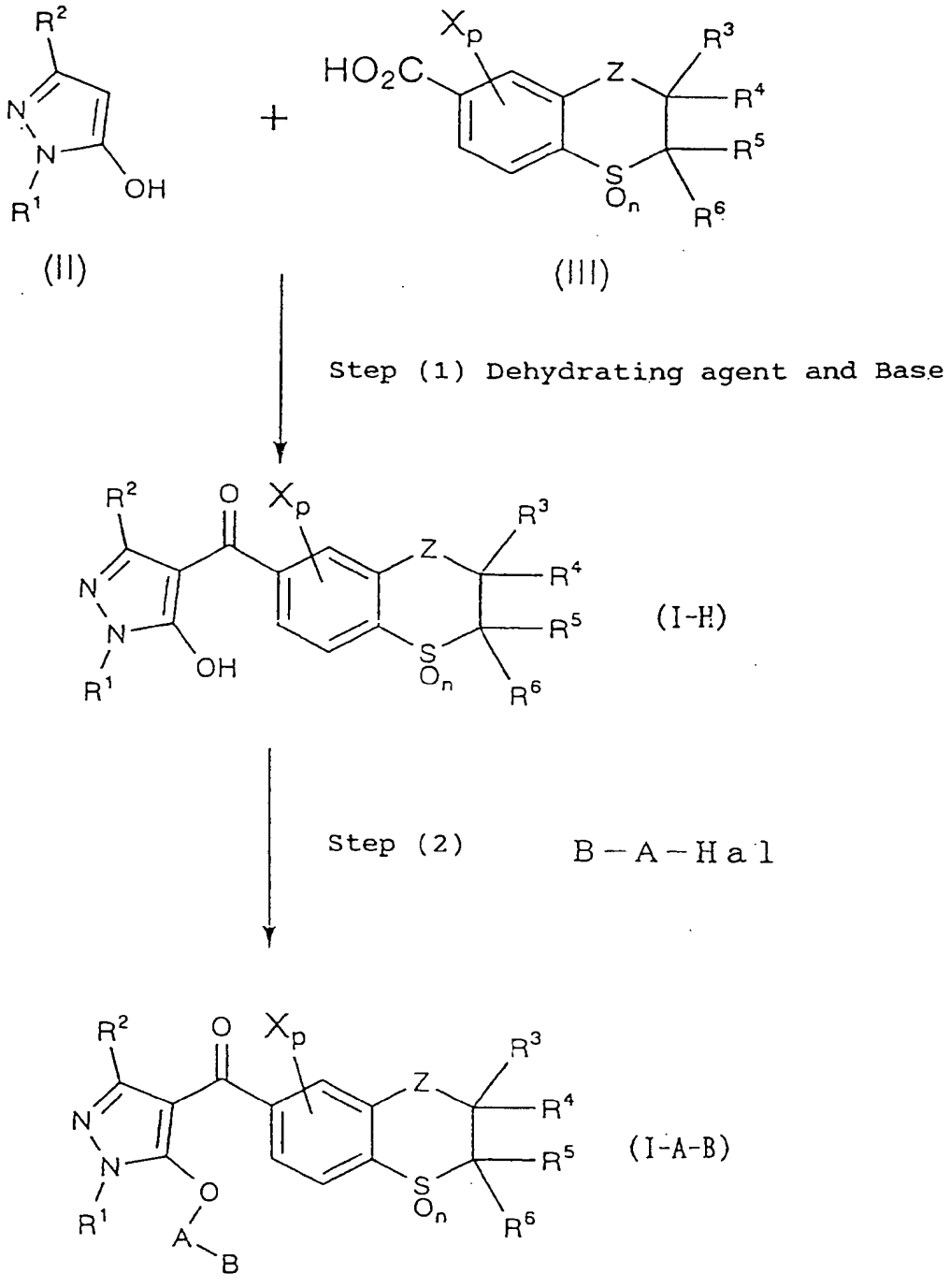
For improving the properties of the preparation and enhancing the herbicidal efficacy of the preparation, the herbicide composition of the present invention may contain polymer compounds or adjuvants such as sodium alginate, carboxymethyl cellulose, carboxyvinyl polymer, gum arabic and hydroxypropylmethylcellulose.

The herbicide composition of the present invention can simultaneously control gramineous weeds and broad-leaved weeds at a low dosage without damaging useful crops (i.e., without phytotoxicity) by soil treatment or foliar treatment for upland crops such as corn, Indian millet and the like before or after the germination of the weeds.

Further, the herbicide composition of the present invention exhibits excellent herbicidal efficacy against weeds in orchards and non-agricultural land (factory field, railroad site, roadside, river bed and fallow field) by soil treatment or foliar treatment.

The herbicide composition of the present invention is applied in an amount, as active ingredients, of approximately 10 to 1,000 g, preferably 70 to 700 g, per hectare. When it is applied to plant stalks and leaves, it is diluted to approximately 100 to 100,000 ppm, preferably 250 to 50,000 ppm, before use.

The pyrazole derivative of the general formula (I) can be produced by the following production process.



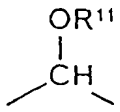
In the above reaction scheme, R^1 ~ R^6, X, p, n ,A, B and Z are as defined in the general formula (I), and Hal is a halogen atom.

Some of thiochromancarboxylic acids of the general formula (III) used as a starting material for the production of the pyrazole derivative of the general formula (I) are already known, and the process for the production thereof is described in International Laid-open Patent Publications WO93/18031, WO94/01431 and WO95/04054. Those which are not known can be produced according to the process described in the above International Laid-open Patent Publications or U. S. Patent 5,035,793.

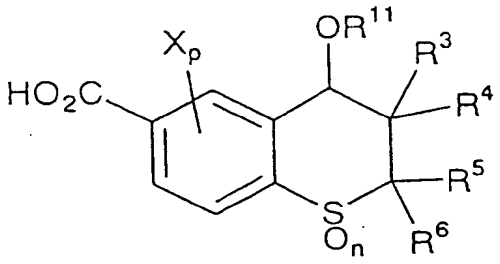
Further, the thiochromancarboxylic acid of the general formula (III) can be produced by one of the following produc-

tion schemes depending upon the structure of substituent. In the reaction schemes, R^1 to R^{12}, X, p, n, A, B and Z are as defined in the general formula (I), Hal is a halogen atom, and q is 1 or 2.

The thiochromancarboxylic acid of the general formula (III) in which Z is

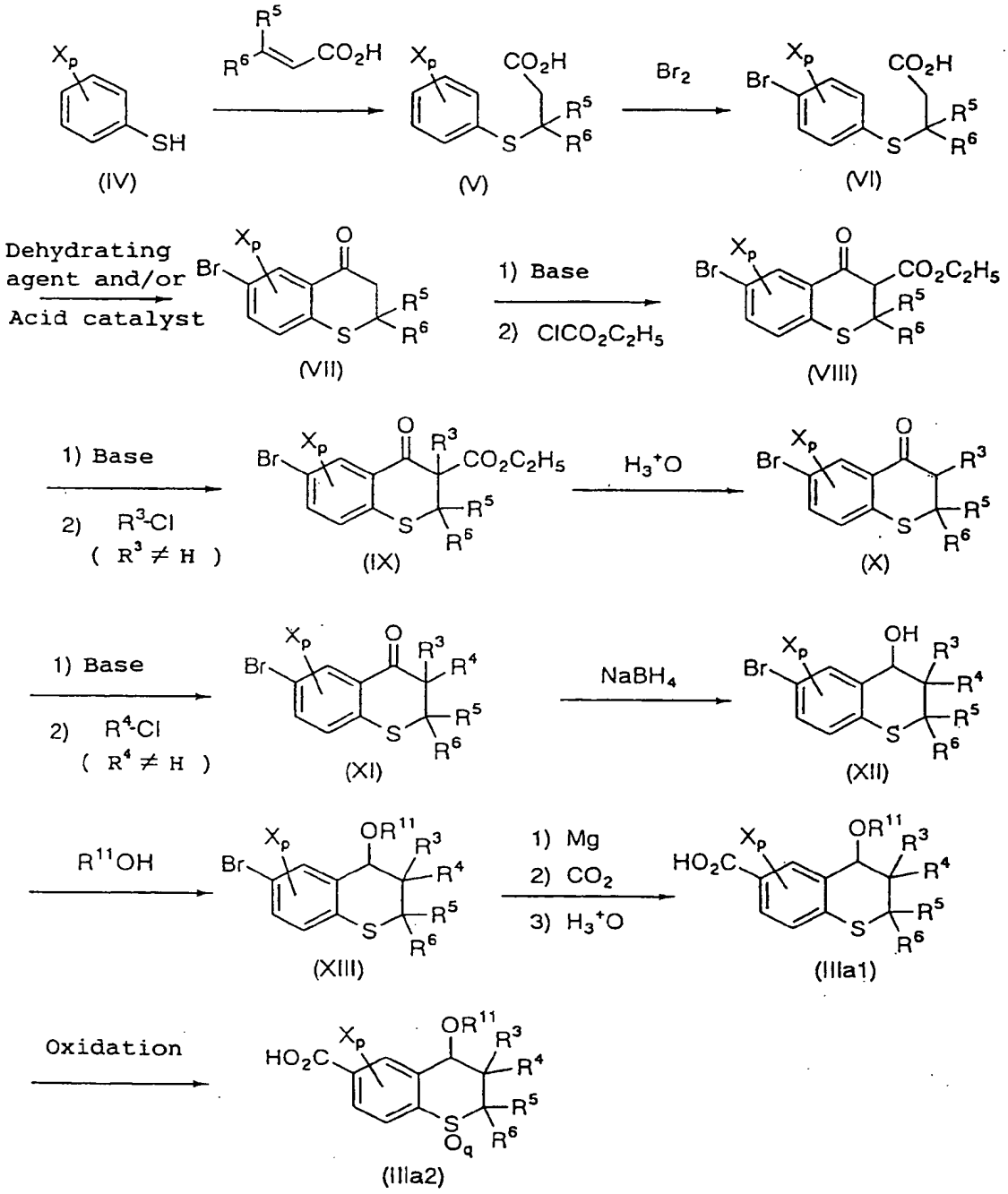


i.e., a thiochromancarboxylic acid of the following general formula (IIIa), can be produced by the method in any one of the production schemes 1 ~ 3.



(IIIa)

Production scheme 1



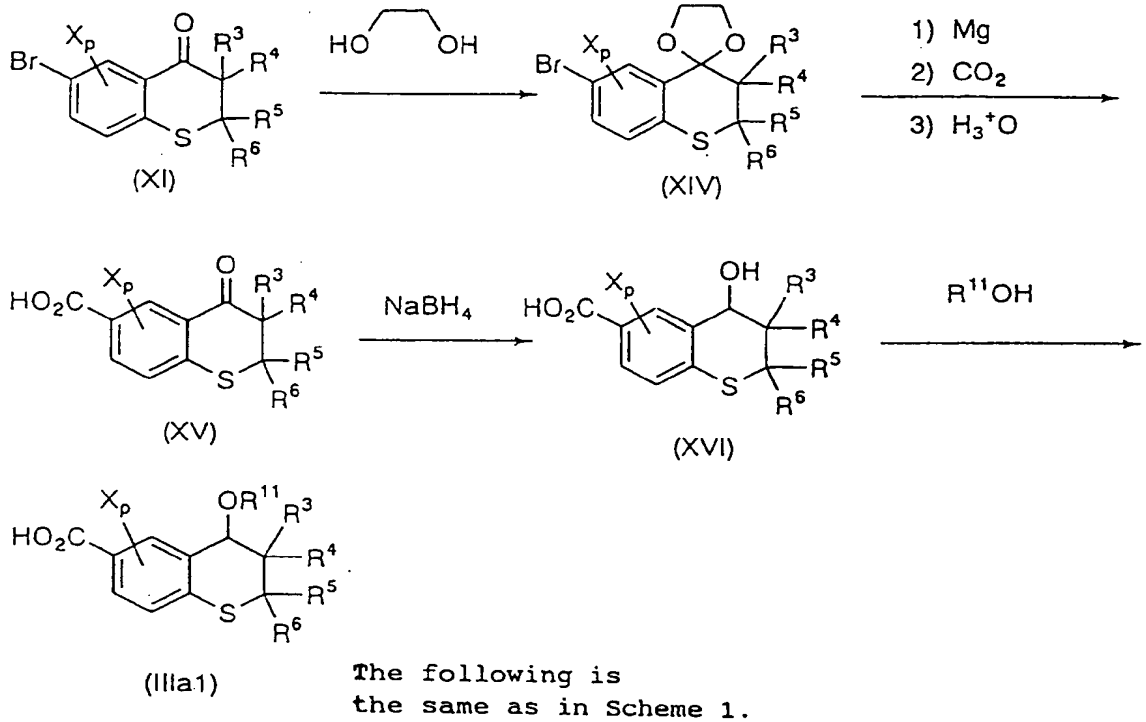
In the production scheme 1, a compound (IIIa1) is a thiochromancarboxylic acid of the general formula (IIIa) in which

n = 0.

A compound (IIIa2) is a thiochromancarboxylic acid of the general formula (IIIa) in which

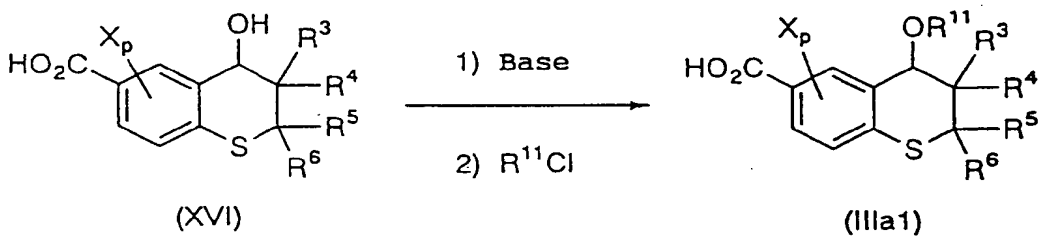
n = 1 or 2.

Production scheme 2

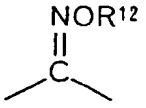


The following is the same as in Scheme 1.

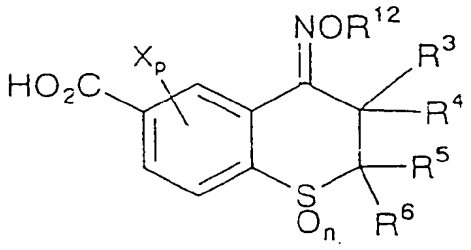
Production scheme 3



To be followed in the same manner as in Scheme 1.
The compound of the general formula (III) in which Z is

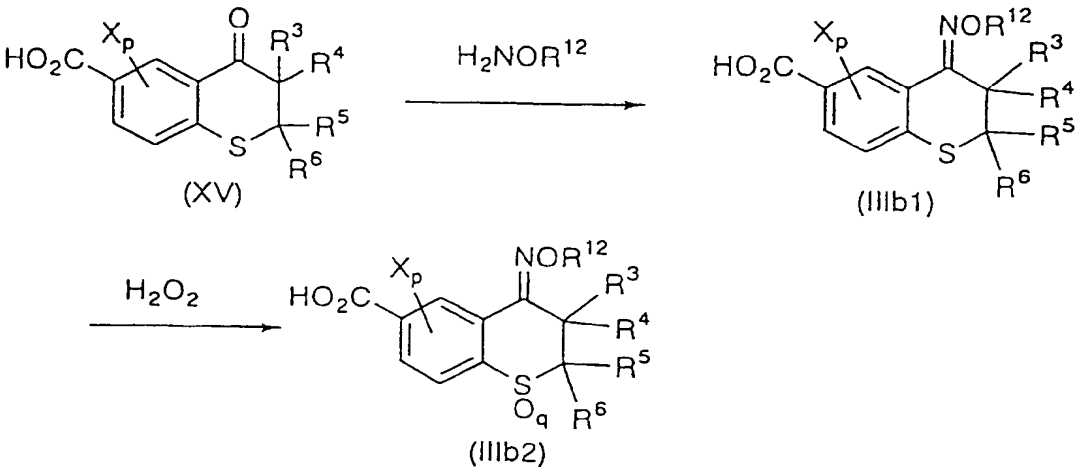


i.e., a thiochromancarboxylic acid of the following general formula (IIIb), can be produced by the method in any one of the production schemes 4 ~ 5.



(IIIb)

Production scheme 4



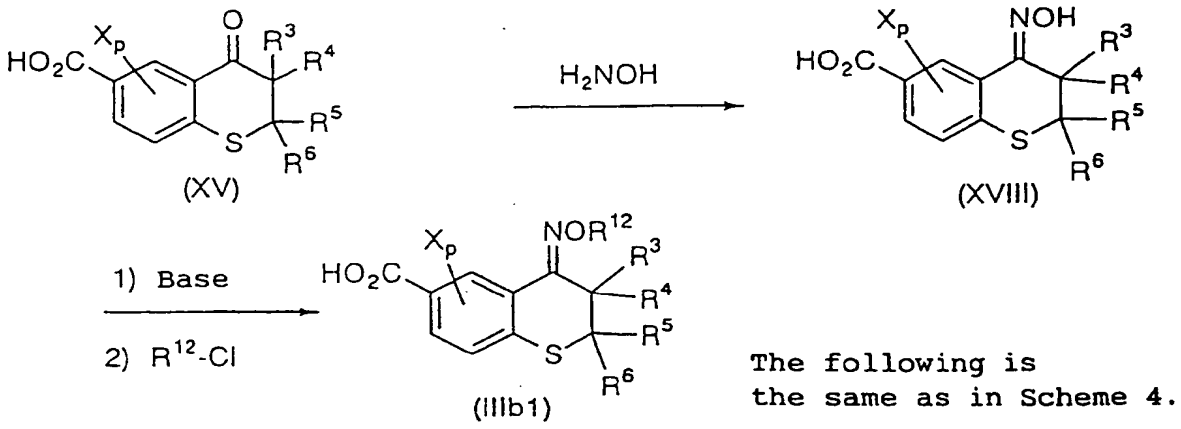
In the production scheme 4, a compound (IIIb1) is a thiochromancarboxylic acid of the general formula (IIIb) in which

n = 0.

A compound (IIIb2) is a thiochromancarboxylic acid of the general formula (IIIb) in which

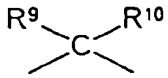
n = 1 or 2.

Production scheme 5

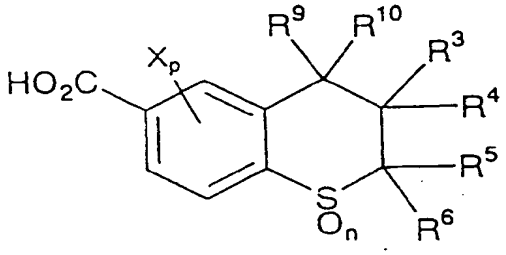


The following is the same as in Scheme 4.

The thiochromancarboxylic acid of the general formula (III) in which Z is



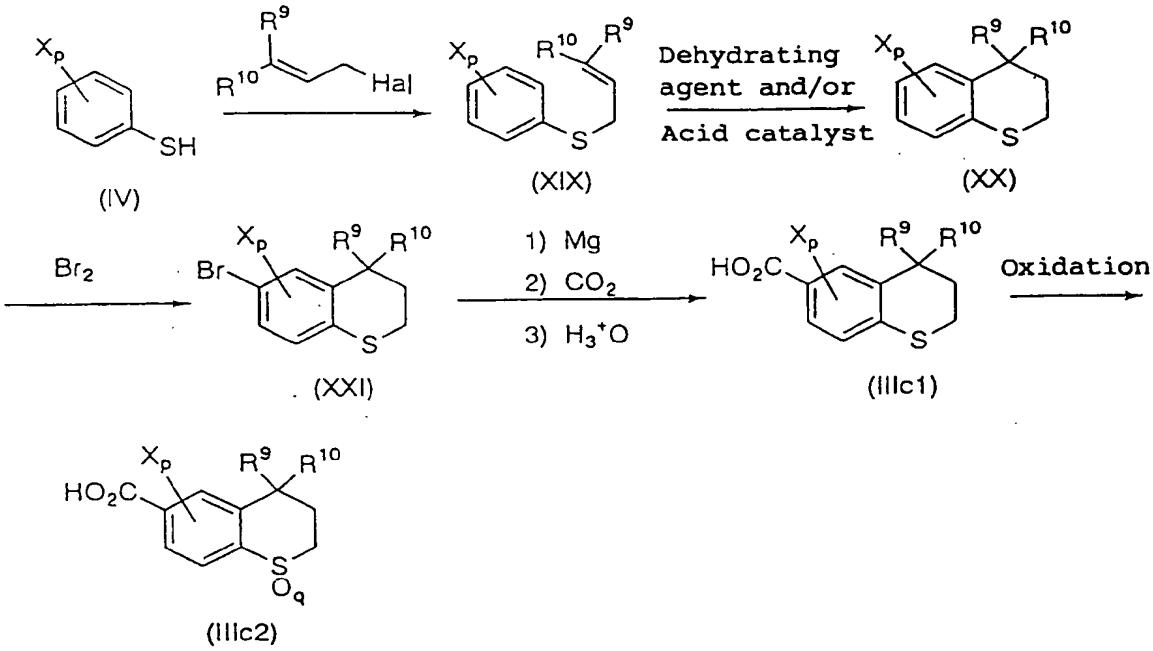
i.e., a thiochromancarboxylic acid of the following general formula (IIIc), can be produced by the method in any one of the production schemes 6 ~ 12.



(IIIc)

Production scheme 6

The production scheme 6 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which R^3, R^4, R^5 and R^6 are all hydrogen atoms.



In the production scheme 6, a compound (IIIc1) is a thiochromancarboxylic acid of the general formula (IIIc) in which

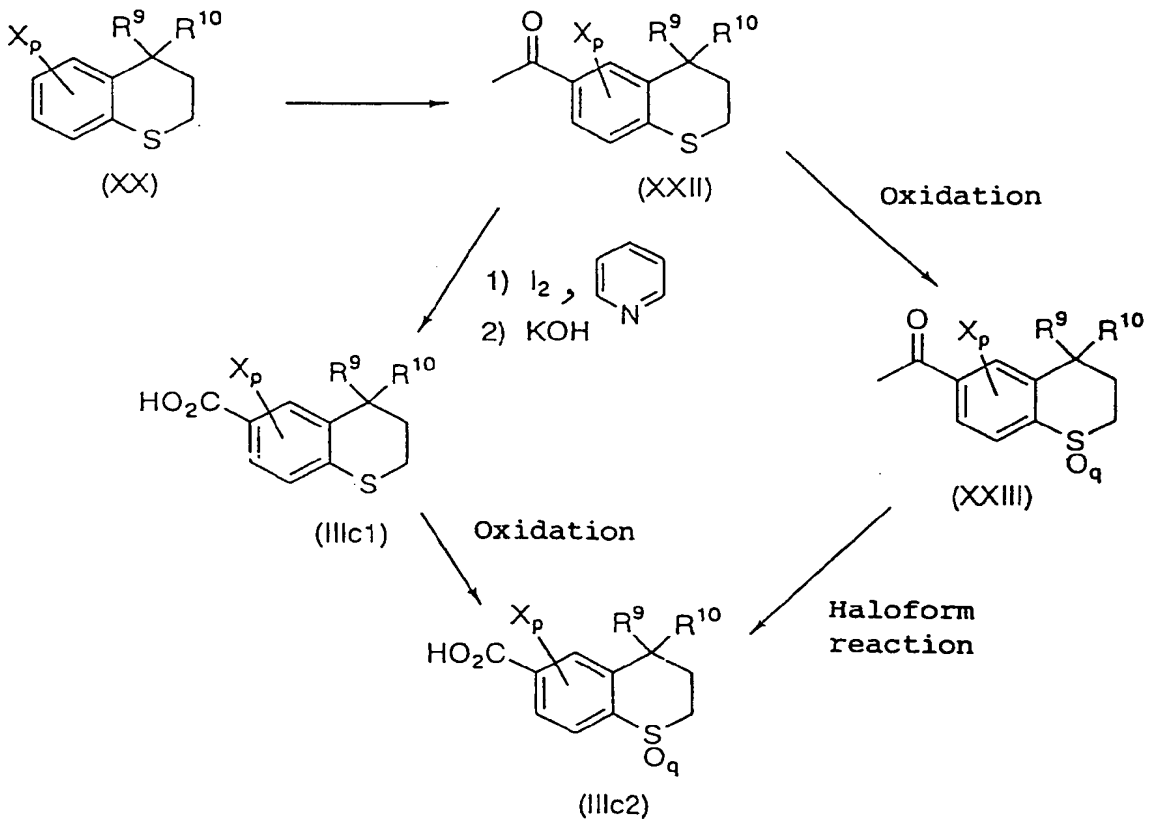
$R^3 = R^4 = R^5 = R^6$ = hydrogen atom, and
n = 0 (sulfide).

A compound (IIIc2) is a thiochromancarboxylic acid of the general formula (IIIc) in which

$R^3 = R^4 = R^5 = R^6$ = hydrogen atom, and
n = 1 (sulfoxide) or 2 (sulfone).

Production scheme 7

The thiochromancarboxylic acid of the general formula (IIIc1) or (IIIc2) in the above production scheme 7 can be also produced by the production scheme 7.

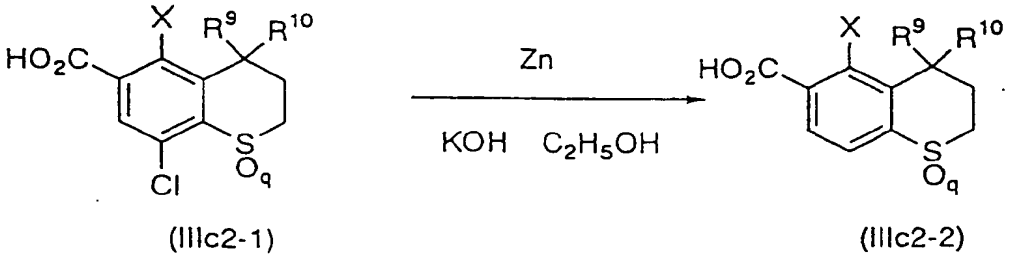


Production scheme 8

A thiochromancarboxylic acid of the general formula (IIIc2) in which

p = 1 and
Position of substituent X = 5-position on the thiochroman ring,

can be also produced by the production scheme 7.

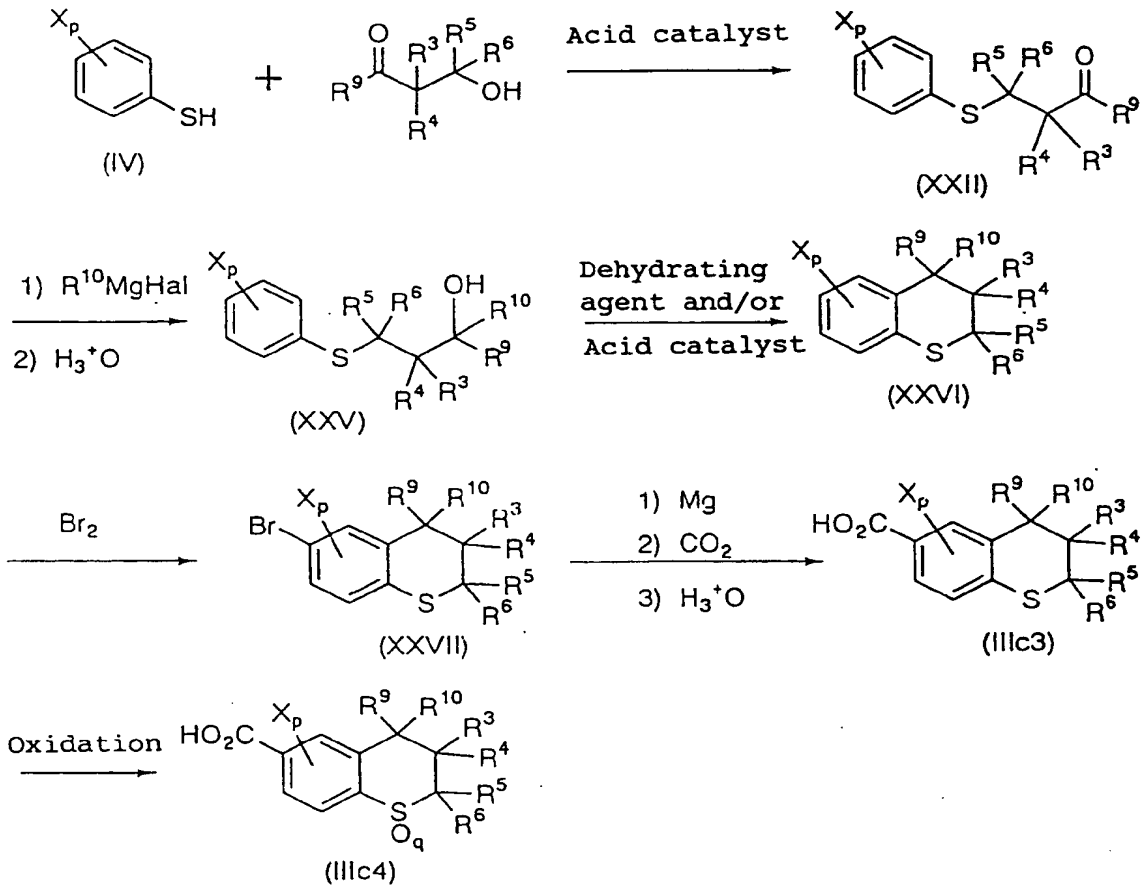


Production scheme 9

The production scheme 9 is directed to a process for the production of a thiochromancarboxylic acid of the general

formula (IIIc) in which

R^{10} = C_1~C_4 alkyl group or C_2~C_5 alkenyl group and
R^9 = C_1~C_4 alkyl group or C_1~C_4 haloalkyl group.



(IV) (XXII) (XXV) (XXVI) (XXVII) (IIIc3) (IIIc4)

In the production scheme 9, a compound (IIIc3) is a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = C_1~C_4 alkyl group or C_2~C_5 alkenyl group,
R^9 = C_1~C_4 alkyl group or C_1~C_4 haloalkyl group, and
n = 0.

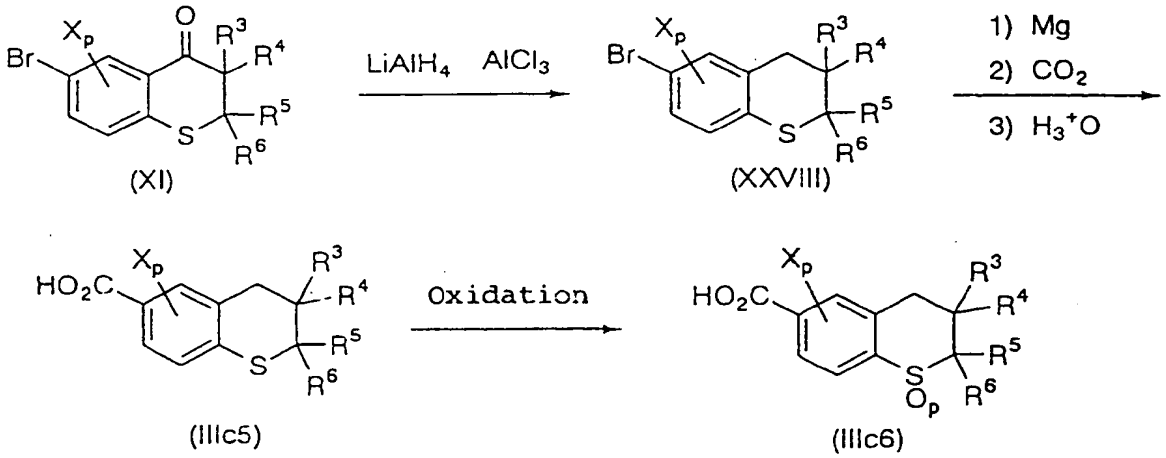
A compound (IIIc4) is a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = C_1~C_4 alkyl group or C_2~C_5 alkenyl group,
R^9 = C_1~C_4 alkyl group or C_1~C_4 haloalkyl group, and
n = 1 or 2.

Production scheme 10

The production scheme 10 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which

R^9 = R^{10} = hydrogen atom.



(XI) (XXVIII) (IIIc5) (IIIc6)

In the production scheme 10, a compound (IIIc5) is a compound of the general formula (IIIc) in which

$R^9 = R^{10}$ = hydrogen atom, and
n = 0.

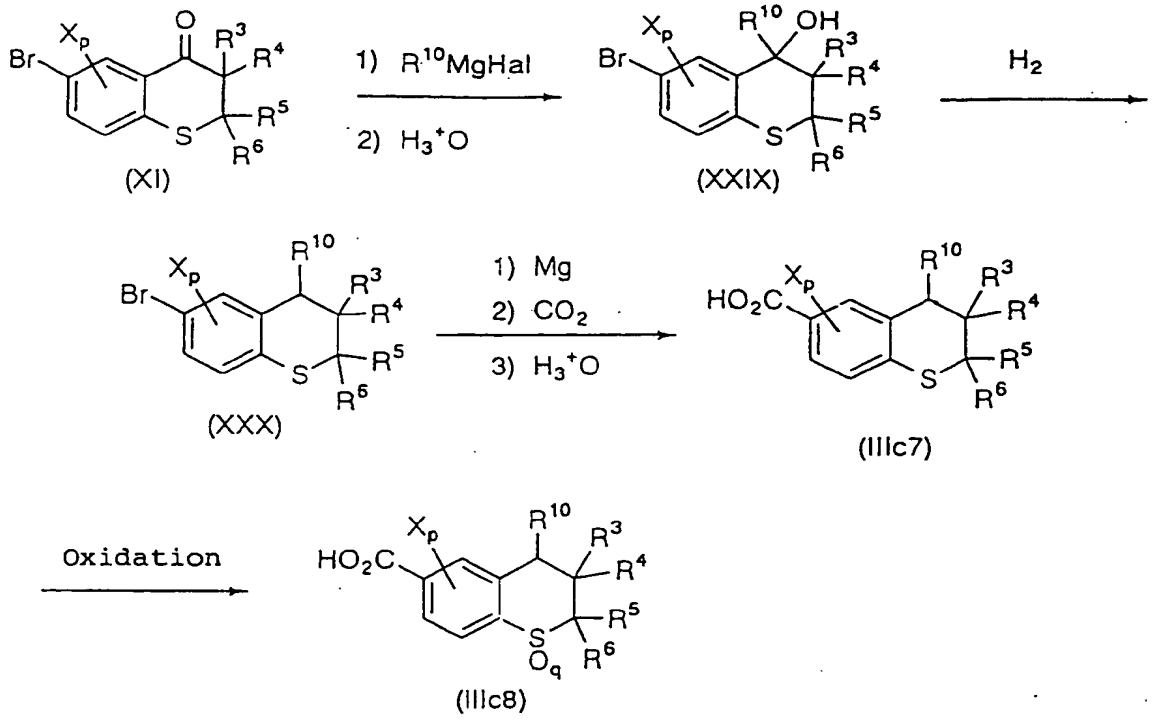
A compound (IIIc6) is a compound of the general formula (IIIc) in which

$R^9 = R^{10}$ = hydrogen atom, and
n = 1 or 2.

Production scheme 11

The production scheme 11 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which

R^9 = hydrogen atom.



In the production scheme 11, a compound (IIIc7) is a compound of the general formula (IIIc) in which

R^9 = hydrogen atom, and
n = 0.

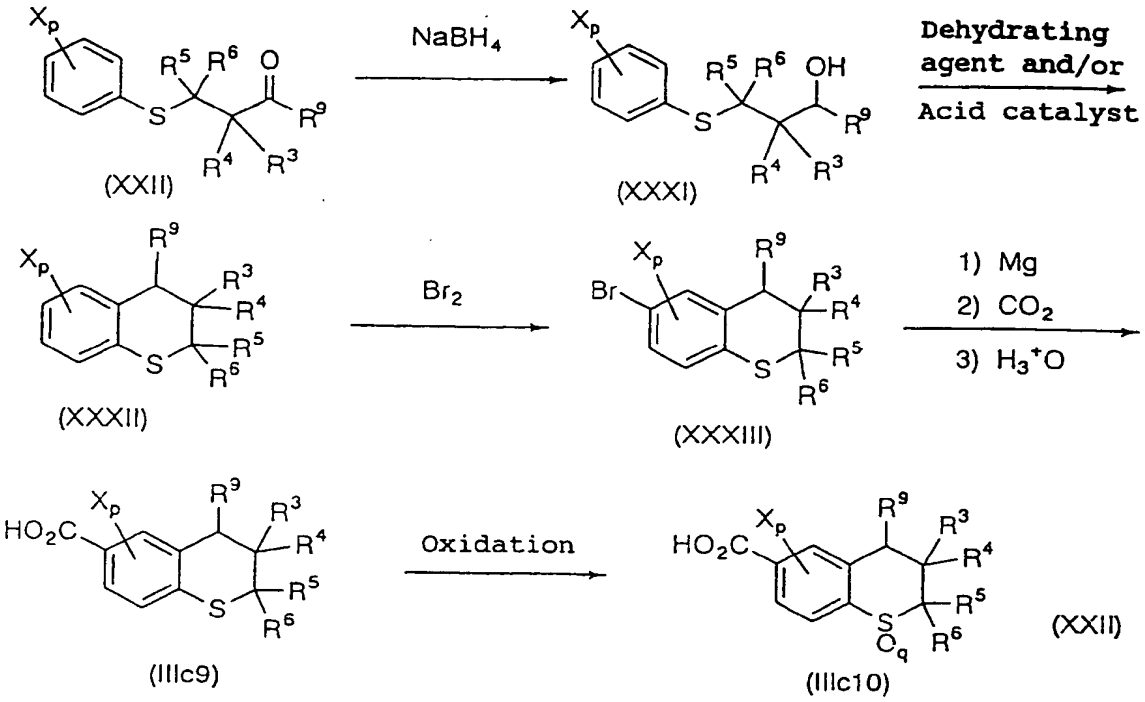
A compound (IIIc8) is a compound of the general formula (IIIc) in which

R^9 = hydrogen atom, and
n = 1 or 2.

Production scheme 12

The production scheme 12 is directed to a process for the production of a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = hydrogen atom.



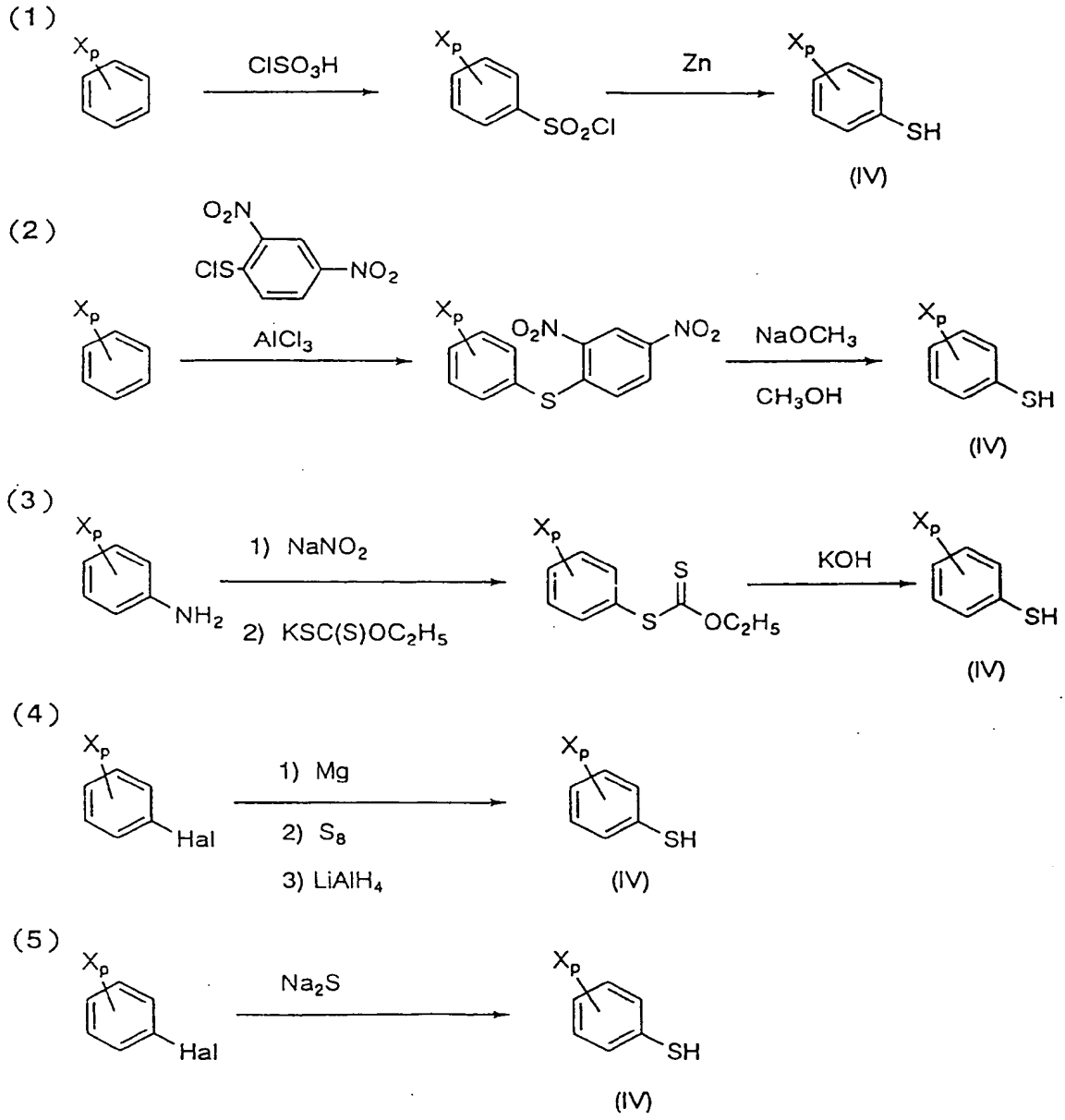
In the production scheme 12, a compound (IIIc9) is a thiochromancarboxylic acid of the general formula (IIIc) in which

R^{10} = hydrogen atom, and
n = 0.

A compound (IIIc10) is a compound of the general formula (IIIc) in which

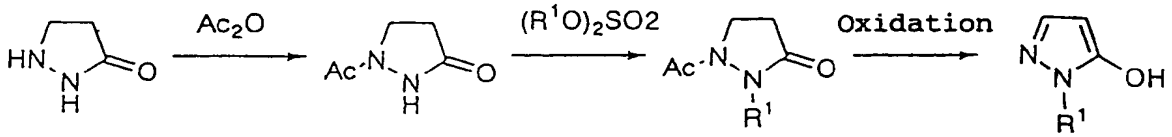
R^{10} = hydrogen atom, and
n = 1 or 2.

Triphenols of the general formula (IV) (production scheme 1), as a starting material for the production of the thiochromancarboxylic acid, can be produced by one of the following methods depending upon their substituents. In the following reaction scheme, X and p are as defined in the general formula (I) and Hal is a halogen atom.

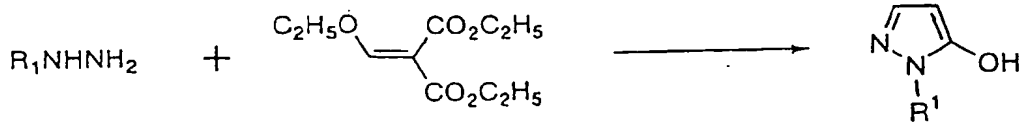


5-Hydroxypyrazoles of the general formula (II), as a starting material for the production of the pyrazole derivative (I) of the present invention, can be produced by one of the following methods depending upon their substituents. In the following reaction schemes, R^1 and R^2 are as defined in the general formula (I).

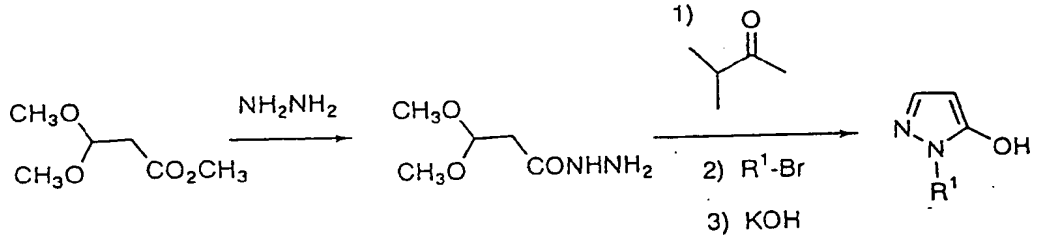
(1) Process mentioned in East German Pat. No. 83145



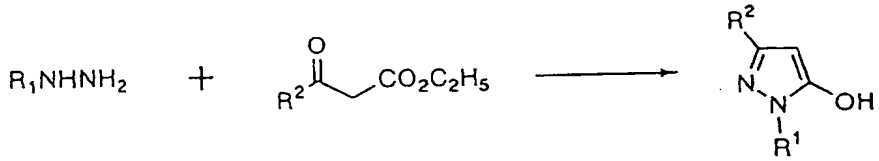
(2) Process mentioned in US Pat. No. 4744815



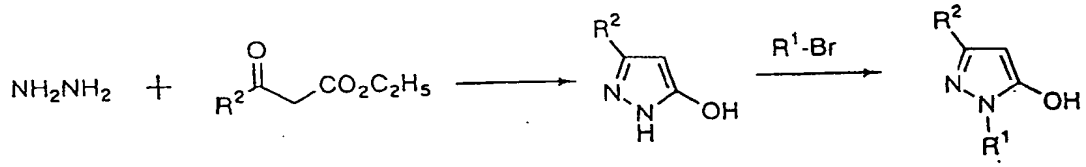
(3) Process mentioned in Japanese Laid-open No. Hei-3-44375



(4)



(5)



The above (1)~(3) are directed to a process for the production of 5-hydroxypyrazoles of the general formula (II) in which

R^2 = hydrogen atom.

The above (5) ~ (6) are directed to a process for the production of 5-hydroxypyrazoles of the general formula (II) in which

R^2 = C_1~C_4 alkyl group, C_1~C_4 haloalkyl group or C_2~C_4 alkoxy group.

Examples

The present invention will be specifically explained with reference to Production Referential Examples, Preparation Examples and Examples hereinafter.

Production Referential Examples

Production Referential Example 1

Synthesis of 4-methoxy-5-methyl-6-(5-cyclohexylcarbonyloxy-1-ethylpyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ia-7)

As a starting material, 4-methoxy-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide corresponding to pyrazole derivative (I-H) was used. O.4 Gram (1.1 mmol) thereof was dissolved in 4 ml of methylene chloride, and 0.22 g (2.2 mmol) of triethylamine as a base and 0.19 g (1.3 mmol) of cyclohexylcarbonyl chloride corresponding to compound B-A-Hal as a reaction reagent were added. The mixture was allowed to react at room temperature for 8 hours. A saturated sodium carbonate aqueous solution was added to the reaction mixture, and the resultant mixture was extracted with ethyl acetate. An organic layer was dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the resultant oil was purified by flush column chromatography (Wako Gel C-300; hexane/ethyl acetate = 1:1) to give 0.28 g (yield 54 %) of 4-methoxy-5-methyl-6-(5-cyclohexylcarbonyloxy-1-ethylpyrazol-4-yl)carbonylthiochroman-1,1-dioxide.

Referential Production Example 2

Synthesis of 4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-1)

A 100-ml eggplant type flask was charged with 1.1 g (2.9 mmol) of 4-methoxyimino-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide corresponding to pyrazole derivative (I-H) as a starting material, and 20 ml of methylene chloride was added and allowed to dissolve it. Then, a solution of 0.41 g of potassium carbonate in 20 ml of distilled water was added. Further, a solution of 0.6 g (4.2 mmol) of n-propanesulfonyl chloride corresponding to compound B-A-Hal as a reaction reagent in 5 ml of methylene chloride was added, and further, 0.05 g of benzyltriethylammonium chloride as a catalyst was added. The mixture was allowed to react at room temperature for 24 hours with stirring. After the reaction was completed, a methylene chloride layer was separated and dried over anhydrous sodium sulfate, and then the methylene chloride was distilled off under reduced pressure. The resultant oily substance was purified with a column packed with silica gel. A mixture of ethyl acetate with n-hexane was used as a developer solution.

By the above procedures, 0.88 g of 4-methoxyimino-5-methyl-6-(1-ethyl-5-n-propylsulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-1) was obtained as a solid. The yield thereof was 62 %.

Referential Production Example 3

Synthesis of 4-methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-2)

0.4 Gram (1.1 mmol) of 4-methoxyimino-5-methyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide corresponding to pyrazole derivative (I-H), 0.23 g (1.2 mmol) of phenacyl bromide corresponding to compound B-A-Hal and 0.15 g of potassium carbonate were added to 10 ml of acetone, and the mixture was stirred under heat for 8 hours. Insolubles were removed by filtration, and then the acetone was distilled off. The residue was dissolved in ethyl acetate, and the mixture was washed with a saturated sodium chloride aqueous solution and dried over sodium sulfate. The ethyl acetate was distilled off under reduced pressure, and the residue was subjected to column chromatography (hexane/ethyl acetate) to give 4-methoxyimino-5-methyl-6-(1-ethyl-5-phenacyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Copound Ib-2) at a yield of 52 %.

Referential Production Example 4

Synthesis of 4-methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ib-3)

4-Methoxyimino-5-methyl-6-(1-ethyl-5-p-toluenesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Com-

pound Ib-3) was obtained in the same manner as in Referential Production Example 2 except that the reaction reaent was replaced with p-toluenesulfonyl chloride corresponding to compound B-A-Hal.

Referential Production Example 5

Synthesis of 4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ic-1)

7.4 Grams (0.026 mol) of 4,4,5,8-tetramethylthiochroman-6-carboxylic acid-1,1-dioxide corresponding to thiochroman carboxylic acid (IIIc), 3.4 g (0.03 mol) of 1-ethyl-5-hydroxypyrazole corresponding to 5-hydroxypyrazole (II) and 6.22 g (0.03 mol) of DCC (N,N'-dicyclohexylcarbodiimide) were all together added to 50 ml of tert-amyl alcohol, and the mixture was stirred at room temperature for 30 minutes. Then, 1.8 g (0.013 mol) of anhydrous potassium carbonate was added. The reaction mixture was allowed to react at 80°C for 8 hours, and the reaction solvent was distilled off under reduced pressure. The resultant residue was dispersed in a 5 % potassium carbonate aqueous solution and ethyl acetate to separate it into two layers. Further, the aqueous layer was adjusted to a pH of 1 with 5 % hydrochloric acid, and the formed solid was recovered by filtration to give 6.13 g (yield 62 %) of 4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ic-1).

Referential Production Example 6

Synthesis of 4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound 1c-2)

0.7 Gram (1.9 mmol) of the 4,4,5,8-tetramethyl-6-(1-ethyl-5-hydroxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ic-1) corresponding to pyrazole derivative (I-H), obtained in Referential Production Example 5, was dissolved in 8 ml of methylene chloride. Then, a solution of 0.51 g (3.8 mmol) of potassium carbonate in 5 ml of water was added, and further, 0.49 g (3.8 mmol) of ethanesulfonyl chloride and 0.05 g (0.2 mmol) of benzyltriethylammonium chloride, corresponding to compound B-A-Hal, were added. The mixture was allowed to react at room temperature for 2 hours, and further refluxed under heat for 2 hours. The reaction mixture was allowed to cool, and thenamethylene chloride layer was recovered and dried over anhydrous sodium sulfate. The solvent was distilled off under reduced pressure, and the resultant oil was purified by silica gel column chromatography to give 0.73 g (yield 82 %) of 4,4,5,8-tetramethyl-6-(1-ethyl-5-ethanesulfonyloxypyrazol-4-yl)carbonylthiochroman-1,1-dioxide (Compound Ic-2).

Table 1 shows physical property data of the compounds obtained in the above Referential Production Examples 1 ~ 6.

Table 1

Prepn Ex. No.	Compd No.	N.M.R.(ppm) Internal standard: tetramethylsilane Solvent: deuterochloroform	I.R. (cm^{-1}) KBr tablet	mp (°C)
1	Ia-7	1.42(3H,t,J=7.3Hz) 1.3-2.0(10H,m) 2.33(3H,s)2.3-2.8(3H,m) 3.1-3.3(1H,m)3.47(3H,s) 3.6-3.8(1H,m) 3.99(2H,q,J=7.3Hz) 4.52(1H,t,J=2.9Hz) 7.44(1H,d,J=8.2Hz)7.61 (1H,s) 7.86(1H,d,J=8.2Hz)	2970,1800, 1670,1300, 1140	139.0-141.0
2	Ib-1	1.18(3H,t)1.52(3H,t)2.00 2.20(2H,m) 2.52(3H,s)3.35 (4H,t)3.73(2H,t 4.06(3H,s)4.23(2H,q) 7.45(H,s 7.48(H,d)7.96(H,d)	3000,2960, 1665, 1135,1325, 1190,1395	146.0-150.7
3	Ib-2	1.51(3H,t)2.41(3H,s) 3.20-3.40(4H,m) 4.02(3H,s)4.28(2H,q) 6.19(2H,s) 7.19(H,s)7.30-8.10(7H,m)	2950,1710, 1650,1320, 1130	-----
4	Ib-3	1.49(3H,t)2.47(3H,s) 2.49(3H,s 3.3-3.5(4H,m)4.05 (3H,s)4.17(2H,q) 7.35(1H,s)7.4-8.0(6H,m)	2950,1680, 1320,1130	glass -like
5	Ic-1	1.45(3H,t)1.55(6H,S) 2.30-2.50(2H,m) 2.50(3H,s)2.80(3H,s) 3.40-3.60(2H,m) 4.10(2H,q)6.20(H,s) 7.20(H,s)	2550-3500, 2950,3000, 1630,1290, 1130	208.8-209.3
6	Ic-2	1.50(3H,t)1.60(6H,s) 1.70(3H,t) 2.30-2.60(2H,m)2.50(3H,s) 2.80(3H,s) 3.30-3.60(2H,m)3.80(2H,q) 4.20(2H,q) 7.10(H,s)7.40(H,s)	2940,3000, 1660,1180, 1140,1290, 1380	164.1-165.7

Preparation Examples

The method of forming preparations will be explained with reference to Preparation Examples hereinafter. "Part" in the following Preparation Examples stands for part by weight.

Preparation Example 1 [Wettable powder]

Compound (Ia-1), (Ib-1) or (Ic-1)	5 parts
Compound (B-1)	25 parts
Diatomaceous earth	52 parts
White carbon	15 parts
Sodium alkylbenzenesulfonate	2 parts
Sodium ligninsulfonate	1 part

The above components were mixed and homogeneously mixed and pulverized to give 100 part of a wettable powder.

Preparation Example 2 [Emulsifiable concentrate]

Compound (Ia-1), (Ib-1) or (Ic-1)	5 parts
Compound (B-2)	25 parts
Xylene	30 parts
Methylnaphthalene	20 parts
Sorpol 2680 (Surfactant supplied by Toho Chemical Co., Ltd.)	20 parts

The above components were homogeneously dissolved and mixed to give 100 parts of an emulsifiable concentrate.

Preparation Example 3 [Dust]

Compound (Ia-1), (Ib-1) or (Ic-1)	0.3 part
Compound (B-3)	1.7 part
Diatomaceous earth	20 parts
Talc	78 parts

The above components were mixed and homogeneously mixed and pulverized to give 100 parts of a dust.

Preparation Example 4 [Flowable preparation]

Compound (Ia-1), (Ib-1) or (Ic-1)	4 parts
Compound (B-4)	25 parts
Methyl cellulose	0.3 part
Colloidal silica	1.5 part
Sodium ligninsulfonate	1 part
Polyoxyethylene nonylphenyl ether	2 parts
Water	66.2 parts

The above components were fully mixed and dispersed, and the resultant mixture in a slurry state was wet-pulverized to give 100 parts of a stable flowable preparation.

Preparation Example 5 [Wettable powder]

97 Parts of clay (trade name: Zeaklite, supplied by Zeaklite Industry Co., Ltd.) as a carrier, 1.5 parts of alkylarylsulfonate (tradename: Neoplex, supplied by Kao-Atlas K.K.) as a surfactant and 1.5 parts by weight of a nonionic and anionic surfactant (trade name: Sorpol 800A, supplied by Toho Chemical Co., Ltd.) were homogeneously pulverized and mixed to prepare 90 parts of a carrier for a wettable powder. 10 Parts of Compound (Ia-1) ~ (Ia-5), (Ia-7), (Ib-1) ~ (Ib-3) or (Ic-1) or 10 parts of one of Compounds (B-1) ~ (B-2) was homogeneously mixed with, and pulverized together with, the above carrier to give wettable powders. A wettable powder containing one of (Ia-1) ~ (Ia-5), (Ia-7), (Ib-1) ~ (Ib-3) and (Ic-1) and a wettable powder containing one of Compound (B-1) ~ (B-20) were mixed in a predetermined mixing ratio (active ingredient ratio) to give wettable powders.

A biological test of the herbicide composition of the present invention will be shown as Examples hereinafter.

Example 1 [Foliar treatment test]

Seeds of weeds such as cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass and seeds of corn were sown in 1/2,000-are Wagner pots filled with upland soil, and covered with upland soil. Then, the seed were grown in a greenhouse, and when these weeds were at a 1.5 ~ 2.5 leaves stage, a predetermined amount of the herbicide obtained in the above Preparation Example 5 was suspended in water and uniformly sprayed onto foliar portions in a solution amount of 1,000 liters/hectare. Thereafter, the weeds were grown in a greenhouse, and 20 days after the treatment, the herbicide was determined for phytotoxicity to crops and herbicidal efficacy according to the following standard.

The herbicidal efficacy (weed control ratio %) was determined by measuring an above-ground green forage weight in a chemical-treated plot and an above-ground green forage weight in a non-treated plot and applying the measurement values to the following equation (A).

$$\text{Weed control ratio (\%)} = (1 - \text{above-ground green forage weight in treated plot} / \text{above-ground green forage weight in non-treated plot}) \times 100$$

The phytotoxicity was evaluated on the basis of the following six ratings.

Degrees of phytotoxicity

0 - No phytotoxicity to crops is observed.
1 - There is almost no phytotoxicity to crops.
2 - Phytotoxicity to crops is observed to some extent.
3 - Phytotoxicity to crops is observed.
4 - Phytotoxicity to crops is markedly observed.
5 - Crops almost died.

In a foliar treatment test, a herbicide is applied by spraying, and it is therefore difficult to bring the amount of the

herbicide to test weeds and crop into a completely constant value. Further, in both a foliar treatment test and a soil treatment test, it is also difficult to constantly bring test conditions such as temperature, humidity, hours of sunlight or water content in soil into completely constant values in the tests even if the tests are carried out in a greenhouse.

Therefore, the herbicidal efficacy of the single active ingredient of each of Compounds (B-1) ~ (B-20) varied in each test. However, the herbicidal efficacy (synergistic effect) of each herbicide composition of the present invention was evaluated on the basis of the results of the tests on single active ingredients carried concurrently, and was reliable.

Table 2 shows the foliar treatment test results of single active ingredients of Compounds (Ia-1), (Ia-2), (Ia-3), (Ia-4), (Ia-5) and (Ia-7) and Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17).

Tables 3 ~ 8 show the foliar treatment test results of herbicide compositions of Compounds (Ia-1), (Ia-2), (Ia-3), (Ia-4), (Ia-5) and (Ia-7) and Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17).

Table 2

Foliar treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ia-1)	40	80	20	50	60	80	60	0
	20	80	20	30	40	60	30	0
(Ia-2)	40	50	50	30	40	60	50	0
	20	30	50	10	20	40	30	0
(Ia-3)	40	50	60	30	50	60	40	0
	20	30	40	10	30	40	30	0
(Ia-4)	40	50	50	30	40	50	40	0
	20	40	30	10	20	30	20	0
(Ia-5)	40	60	50	30	40	50	30	0
	20	40	30	10	20	30	20	0
(Ia-7)	40	40	50	40	50	50	40	0
	20	30	30	10	20	20	20	0
(B-1)	250	0	0	60	0	0	0	0
(B-10)	250	90	40	30	0	0	0	0
(B-7)	250	80	40	40	0	0	0	0
(B-6)	250	90	0	0	0	0	0	0
(B-20)	250	0	10	0	0	0	0	0
(B-3)	62	90	90	60	0	40	0	0
(B-2)	125	80	0	0	40	60	0	0
(B-4)	125	20	0	0	20	60	0	0
(B-9)	250	100	20	20	0	0	0	0
(B-14)	16	20	20	20	60	50	60	0
(B-15)	10	20	20	20	50	50	50	0
(B-16)	36	0	0	20	80	70	80	0
(B-17)	16	30	20	20	20	20	10	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Table 3 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-1)+(B-1) 40 + 250	100	80	20	90	20	70	100	80	20	80	60	20	100	80	20	100	60	40	0
(Ia-1)+(B-10) 20 + 250	100	98	2	100	52	48	80	51	29	80	40	40	90	60	30	80	30	50	0
(Ia-1)+(B-7) 40 + 250	100	96	4	70	52	18	100	70	30	90	60	30	100	80	20	80	60	20	0
(Ia-1)+(B-6) 40 + 250	100	98	2	80	20	60	100	50	50	80	60	20	100	80	20	90	60	30	0
(Ia-1)+(B-20) 40 + 250	100	80	20	90	20	70	60	50	10	80	60	20	100	80	20	90	60	30	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 4 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-2)+(B-1) 20 + 250	100	30	70	100	50	50	90	64	26	90	20	70	100	40	60	90	30	60	0
(Ia-2)+(B-10) 40 + 250	100	95	5	100	70	30	70	51	19	90	40	50	80	60	20	80	50	30	0
(Ia-2)+(B-7) 40 + 250	100	90	10	100	70	30	80	58	22	50	40	10	70	60	10	70	50	20	0
(Ia-2)+(B-6) 40 + 250	100	95	5	100	50	50	90	30	60	80	40	40	100	60	40	90	50	40	0
(Ia-2)+(B-20) 40 + 250	50	50	0	60	55	5	50	30	20	50	40	10	60	60	0	60	50	10	0
(Ia-2)+(B-3) 40 + 62	100	95	5	100	95	5	100	72	28	90	40	50	100	76	24	100	50	50	0
(Ia-2)+(B-2) 20 + 125	100	86	14	80	50	30	90	10	80	90	52	38	100	76	24	80	30	50	0
(Ia-2)+(B-4) 40 + 125	100	60	40	100	50	50	90	30	60	90	52	38	100	84	16	100	50	50	0
(Ia-2)+(B-9) 40 + 250	100	100	0	100	60	40	80	44	36	50	40	10	70	60	10	50	50	0	0
(Ia-2)+(B-14) 40 + 16	80	60	20	60	60	0	50	44	6	80	76	4	80	80	0	80	80	0	0
(Ia-2)+(B-15) 40 + 10	70	60	10	60	60	0	50	44	6	80	70	10	80	80	0	80	75	5	0
(Ia-2)+(B-16) 40 + 36	60	50	10	50	50	0	50	44	6	90	88	2	90	88	2	90	90	0	0
(Ia-2)+(B-17) 40 + 16	80	65	15	60	40	20	50	44	6	60	52	8	70	68	2	60	55	5	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 5 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-3)+(B-1) 20 + 250	100	30	70	100	40	60	90	64	26	90	30	60	100	40	60	90	30	60	0
(Ia-3)+(B-10) 40 + 250	100	100	0	100	76	24	70	51	19	60	50	10	80	60	20	60	40	20	0
(Ia-3)+(B-7) 40 + 250	100	90	10	100	76	24	100	58	42	60	50	10	70	60	10	60	40	20	0
(Ia-3)+(B-6) 40 + 250	100	95	5	100	60	40	100	30	70	80	50	30	80	60	20	90	40	50	0
(Ia-3)+(B-20) 40 + 250	50	50	0	70	64	6	50	30	20	60	50	10	60	60	0	60	40	20	0
(Ia-3)+(B-3) 40 + 62	100	95	5	100	96	4	90	72	18	90	50	40	100	76	24	100	40	60	0
(Ia-3)+(B-2) 20 + 125	100	86	14	90	40	50	100	10	90	90	58	32	100	76	24	100	30	70	0
(Ia-3)+(B-4) 40 + 125	100	60	40	100	60	40	90	30	60	90	60	30	100	84	16	90	40	50	0
(Ia-3)+(B-9) 40 + 250	100	100	0	100	68	32	80	44	36	50	50	0	70	60	10	50	40	10	0
(Ia-3)+(B-14) 40 + 16	70	60	10	70	68	2	50	44	6	80	80	0	80	80	0	80	76	4	0
(Ia-3)+(B-15) 40 + 10	70	60	10	70	68	2	50	44	6	80	75	5	80	80	0	80	70	10	0
(Ia-3)+(B-16) 40 + 36	60	50	10	60	60	0	50	44	6	90	90	0	90	88	2	90	88	2	0
(Ia-3)+(B-17) 40 + 16	80	65	15	70	68	2	50	44	6	60	60	0	70	68	2	50	46	4	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 6 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-4)+(B-1) 20 + 250	100	40	60	100	30	70	90	64	26	90	20	70	100	30	70	100	20	80	0
(Ia-4)+(B-10) 40 + 250	100	100	0	100	70	30	80	51	29	60	40	20	80	50	30	60	40	20	0
(Ia-4)+(B-7) 40 + 250	100	90	10	100	70	30	100	58	42	60	40	20	70	50	20	70	40	30	0
(Ia-4)+(B-6) 40 + 250	100	95	5	100	50	50	100	30	70	80	40	40	80	50	30	80	40	40	0
(Ia-4)+(B-20) 40 + 250	70	50	20	60	55	5	50	30	20	60	40	20	60	50	10	50	40	10	0
(Ia-4)+(B-3) 40 + 62	100	95	5	100	95	5	90	72	18	90	40	50	100	70	30	90	40	50	0
(Ia-4)+(B-2) 20 + 125	100	88	12	90	30	60	100	10	90	90	52	38	100	72	28	100	20	80	0
(Ia-4)+(B-4) 40 + 125	100	60	40	100	50	50	100	30	70	90	52	38	100	80	20	100	40	60	0
(Ia-4)+(B-9) 40 + 250	100	100	0	100	60	40	90	44	48	50	40	10	60	50	10	50	40	10	0
(Ia-4)+(B-14) 40 + 16	70	60	10	70	60	10	50	44	6	80	76	4	80	75	5	80	76	4	0
(Ia-4)+(B-15) 40 + 10	70	60	10	60	60	0	50	44	6	70	70	0	80	75	5	80	70	10	0
(Ia-4)+(B-16) 40 + 36	60	50	10	60	50	10	50	44	6	90	88	2	90	85	5	90	88	2	0
(Ia-4)+(B-17) 40 + 16	80	65	15	70	60	10	60	44	16	60	52	8	70	60	10	50	46	4	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 7 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-5)+(B-1) 20 + 250	100	40	60	100	30	70	90	64	26	90	20	70	100	30	70	90	20	70	0
(Ia-5)+(B-10) 40 + 250	100	100	0	100	70	30	90	51	39	60	40	20	70	50	20	60	30	30	0
(Ia-5)+(B-7) 40 + 250	100	92	8	100	70	30	100	58	42	60	40	20	60	50	10	70	30	40	0
(Ia-5)+(B-6) 40 + 250	100	96	4	100	50	50	100	30	70	70	40	30	80	50	30	80	30	50	0
(Ia-5)+(B-20) 40 + 250	60	60	0	60	55	5	50	30	20	50	40	10	60	50	10	50	30	20	0
(Ia-5)+(B-3) 40 + 62	100	96	4	100	95	5	90	72	18	100	40	60	100	70	30	100	30	70	0
(Ia-5)+(B-2) 20 + 125	100	68	32	100	50	50	90	30	60	90	52	38	100	80	20	100	30	70	0
(Ia-5)+(B-4) 40 + 125	100	68	32	100	50	50	90	30	60	90	52	38	100	80	20	100	30	70	0
(Ia-5)+(B-9) 40 + 250	100	100	0	100	60	40	90	44	46	60	40	20	60	50	10	50	30	20	0
(Ia-5)+(B-14) 40 + 16	70	68	2	70	60	10	50	44	6	80	76	4	80	75	5	80	72	8	0
(Ia-5)+(B-15) 40 + 10	70	68	2	70	60	10	50	44	6	70	70	0	80	75	5	70	65	5	0
(Ia-5)+(B-16) 40 + 36	70	60	10	70	50	20	50	44	6	90	88	2	90	85	5	90	86	4	0
(Ia-5)+(B-17) 40 + 16	80	72	8	70	60	10	70	44	26	60	52	8	70	60	10	50	37	13	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 8 Foliar treatment (combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)												phyto-toxicity
	(a)		(b)		(c)		(d)		(e)		(f)		corn
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	
(Ia-7)+(B-1) 20 + 250	100	30	70	100	30	70	100	64	36	90	20	70	100	20	80	90	20	70	0
(Ia-7)+(B-10) 40 + 250	100	100	0	100	70	30	100	90	58	32	60	50	10	70	50	20	60	40	20	0
(Ia-7)+(B-7) 40 + 250	100	88	12	100	70	30	100	64	36	60	50	10	70	50	20	70	40	30	0
(Ia-7)+(B-6) 40 + 250	100	94	6	100	50	50	100	40	60	80	50	30	80	50	30	70	40	30	0
(Ia-7)+(B-20) 40 + 250	60	40	20	60	55	5	50	40	10	60	50	10	60	50	10	50	40	10	0
(Ia-7)+(B-3) 40 + 62	100	94	6	100	95	5	90	76	14	100	50	50	100	70	30	100	40	60	0
(Ia-7)+(B-2) 20 + 125	100	86	14	100	30	70	100	30	70	100	52	48	100	68	32	90	20	70	0
(Ia-7)+(B-4) 40 + 125	100	52	48	100	50	50	90	40	50	90	60	30	100	80	20	90	40	50	0
(Ia-7)+(B-9) 40 + 250	100	100	0	100	60	40	90	52	38	60	50	10	60	50	10	50	40	10	0
(Ia-7)+(B-14) 40 + 16	60	52	8	70	60	10	60	52	8	80	80	0	80	75	5	80	76	4	0
(Ia-7)+(B-15) 40 + 10	70	52	18	70	60	10	60	52	8	80	50	30	80	50	30	70	70	0	0
(Ia-7)+(B-16) 40 + 36	50	40	10	70	50	20	60	52	8	90	90	0	90	85	5	90	88	2	0
(Ia-7)+(B-17) 40 + 16	80	58	22	80	60	20	70	52	18	60	60	0	70	60	10	50	46	4	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass
Difference (Δ) = Found value(F) - Expected value(E)
A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 3 ~ 8 show that all the compositions containing Compound (Ia-1) and one of Compounds (B-1), (B-6), (B-7), (B-10) and (B-20) in combination and all the compositions containing one of compounds (Ia-2) ~ (Ia-5) and (Ia-7) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing compound (Ia-1) and one of Compounds (B-1), (B-10), (B-6) and (B-20),

the composition containing Compound (Ia-1) and Compound (B-1) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-1) and Compound (B-10) showed a high synergistic effect on velvet leaf and barnyard grass in particular,

the composition containing Compound (Ia-1) and Compound (B-7) showed a high synergistic effect on slender amaranth and green foxtail in particular,

the composition containing Compound (Ia-1) and Compound (B-6) showed a high synergistic effect on velvet leaf and slender amaranth in particular, and

the composition containing Compound (Ia-1) and Compound (B-20) showed a high synergistic effect on velvet leaf in particular.

Further, the compositions containing compound (Ia-1) and compound (B-1), (B-10) or (B-6) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

In the compositions containing Compound (Ia-2) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-2) and compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-2) and Compound (B-10) showed a high synergistic effect on green foxtail in particular,

the composition containing Compound (Ia-2) and Compound (B-7) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-2) and Compound (B-6) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-2) and Compound (B-20) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ia-2) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular

the composition containing Compound (Ia-2) and Compound (B-2) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-2) and Compound (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-2) and Compound (B-9) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-2) and Compound (B-14) showed a high synergistic effect on cocklebur in particular,

the composition containing compound (Ia-2) and Compound (B-15) showed a high synergistic effect on cocklebur and green foxtail in particular,

the composition containing Compound (Ia-2) and Compound (B-16) showed a high synergistic effect on cocklebur in particular, and

the composition containing Compound (Ia-2) and Compound (B-17) showed a high synergistic effect on cocklebur and velvet leaf in particular.

In the compositions containing Compound (Ia-3) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-3) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-10) showed a high synergistic effect on velvet leaf in particular,

the composition containing Compound (Ia-3) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ia-3) and Compound (B-6) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-20) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-4) showed a high synergistic effect on slender amaranth and barnyard grass in particular,

the composition containing Compound (Ia-3) and Compound (B-9) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-3) and Compound (B-14) showed a high synergistic effect on cocklebur

in particular,
the composition containing Compound (Ia-3) and Compound (B-15) showed a high synergistic effect on cocklebur and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-16) showed a high synergistic effect on cocklebur in particular, and
the composition containing Compound (Ia-3) and Compound (B-17) showed a high synergistic effect on cocklebur in particular.

In the compositions containing Compound (Ia-4) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-4) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-10) showed a high synergistic effect on velvet leaf, slender amaranth and crabgrass in particular,
the composition containing Compound (Ia-4) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-4) and Compound (B-6) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-4) and Compound (B-20) showed a high synergistic effect on cocklebur, slender amaranth and green foxtail in particular,
the composition containing Compound (Ia-4) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-9) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ia-4) and Compound (B-14) showed a high synergistic effect on cocklebur in particular,
the composition containing Compound (Ia-4) and Compound (B-15) showed a high synergistic effect on cocklebur and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-16) showed a high synergistic effect on cocklebur and velvet leaf in particular, and
the composition containing Compound (Ia-4) and Compound (B-17) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing Compound (Ia-5) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-5) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-10) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-7) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-20) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-9) showed a high synergistic effect on velvet leaf and slender amaranth in particular,

the composition containing Compound (Ia-5) and Compound (B-14) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ia-5) and Compound (B-15) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ia-5) and Compound (B-16) showed a high synergistic effect on velvet leaf in particular, and
the composition containing Compound (Ia-5) and Compound (B-17) showed a high synergistic effect on slender amaranth in particular.

In the compositions containing Compound (Ia-7) and one of Compounds (B-1) ~ (B-4), (B-6), (B-7), (B-9), (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ia-7) and Compound (B-1) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-10) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-7) and Compound (B-7) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth in particular,
the composition containing Compound (Ia-7) and Compound (B-20) showed a high synergistic effect on cocklebur in particular,
the composition containing Compound (Ia-7) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-2) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-9) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-7) and Compound (B-14) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-7) and Compound (B-15) showed a high synergistic effect on crabgrass in particular,
the composition containing Compound (Ia-7) and Compound (B-16) showed a high synergistic effect on velvet leaf in particular, and
the composition containing Compound (Ia-7) and Compound (B-17) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular.

Table 9 shows the results of foliar treatment tests of single active ingredients of Compounds (Ib-1), (Ib-2), (Ib-3) and Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17).

Tables 10 to 12 show the results of foliar treatment tests of compositions containing one of Compounds (Ib-1), (Ib-2) and (Ib-3) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17).

Table 9

Foliar treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ib-1)	40	60	20	20	40	60	40	0
	20	40	10	20	20	40	20	0
(Ib-2)	40	50	30	30	40	60	40	0
	20	30	20	20	20	40	20	0
(Ib-3)	40	50	30	20	50	60	40	0
	20	40	20	20	20	40	30	0
(B-1)	250	20	0	0	0	0	0	0
(B-2)	125	80	0	0	40	60	0	0
(B-3)	62	90	90	60	0	40	0	0
(B-4)	125	20	0	0	20	60	0	0
(B-5)	62	20	20	0	0	0	0	0
(B-6)	250	20	0	20	0	0	0	0
(B-7)	125	90	90	40	0	0	0	0
(B-8)	250	90	0	0	0	0	0	0
(B-9)	250	90	20	40	0	0	0	0
(B-10)	250	90	80	0	0	20	0	0
(B-14)	16	20	20	20	60	50	60	0
(B-15)	10	20	20	20	50	50	50	0
(B-16)	36	0	0	20	80	70	80	0
(B-17)	16	30	20	20	20	20	10	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Table 10 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ib-1)+(B-1) 20 + 250	80	52	28	100	10	90	60	20	40	100	20	80	100	40	60	80	20	60	0
(Ib-1)+(B-2) 20 + 125	100	88	12	100	10	90	80	20	60	100	52	48	100	76	24	80	20	60	0
(Ib-1)+(B-3) 40 + 62	100	96	4	100	92	8	100	68	32	100	40	60	100	76	24	100	40	60	0
(Ib-1)+(B-4) 40 + 125	100	68	32	100	20	80	80	20	60	90	52	38	100	84	16	100	40	60	0
(Ib-1)+(B-5) 20 + 62	100	52	48	100	28	72	40	20	20	80	20	60	80	40	40	80	20	60	0
(Ib-1)+(B-6) 40 + 250	100	68	32	100	20	80	100	36	64	90	40	50	100	60	40	90	40	50	0
(Ib-1)+(B-7) 40 + 125	100	96	4	100	92	8	100	52	48	40	40	0	60	60	0	40	40	0	0
(Ib-1)+(B-8) 20 + 250	100	90	10	100	20	80	60	20	40	100	40	60	100	60	40	80	40	40	0
(Ib-1)+(B-9) 40 + 250	100	96	4	80	36	44	100	36	64	60	40	20	80	60	20	40	40	0	0
(Ib-1)+(B-10) 40 + 250	100	96	4	100	84	16	40	20	20	90	40	50	90	68	22	80	40	40	0
(Ib-1)+(B-14) 40 + 16	80	68	12	60	36	24	40	36	4	80	76	4	80	80	0	80	76	4	0
(Ib-1)+(B-15) 40 + 10	70	68	2	40	36	4	40	36	4	80	70	10	80	80	0	80	70	10	0
(Ib-1)+(B-16) 40 + 36	70	60	10	40	20	20	40	36	4	90	88	2	90	88	2	90	88	2	0
(Ib-1)+(B-17) 40 + 16	80	72	8	40	36	4	40	36	4	60	52	8	70	68	2	50	48	2	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 11 Foliar treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(1b-2)+(B-1) 20 + 250	90	44	46	100	20	80	60	20	40	90	20	70	100	40	60	90	20	70	0
(1b-2)+(B-2) 20 + 125	100	86	14	90	20	70	90	20	70	90	52	38	100	76	24	80	20	60	0
(1b-2)+(B-3) 40 + 62	100	95	5	100	93	7	100	72	28	100	40	60	100	76	24	100	40	60	0
(1b-2)+(B-4) 40 + 125	100	60	40	100	30	70	80	30	50	100	52	48	90	84	6	100	40	60	0
(1b-2)+(B-5) 20 + 62	100	44	56	90	36	54	40	20	20	80	20	60	90	40	50	80	20	60	0
(1b-2)+(B-6) 40 + 250	100	60	40	90	30	60	100	44	56	90	40	50	100	60	40	90	40	50	0
(1b-2)+(B-7) 40 + 125	100	95	5	100	93	7	90	58	32	60	40	20	80	60	20	60	40	20	0
(1b-2)+(B-8) 20 + 250	100	95	5	100	30	70	60	20	40	90	40	50	90	60	30	90	40	50	0
(1b-2)+(B-9) 40 + 250	100	95	5	90	44	46	100	44	56	60	40	20	80	60	20	40	40	0	0
(1b-2)+(B-10) 40 + 250	100	95	5	100	86	14	50	30	20	90	40	50	90	68	22	90	40	50	0
(1b-2)+(B-14) 40 + 16	60	60	0	50	44	6	50	44	6	80	76	4	80	80	0	80	76	4	0
(1b-2)+(B-15) 40 + 10	70	60	10	50	44	6	70	44	26	80	70	10	80	80	0	80	70	10	0
(1b-2)+(B-16) 40 + 36	60	50	10	40	30	10	50	44	6	90	88	2	90	88	2	90	88	2	0
(1b-2)+(B-17) 40 + 16	80	65	15	50	44	6	50	44	6	60	52	8	70	68	2	50	46	4	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 12 Foliar treatment (combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%) (a) (F)	(E)	(Δ)	(b) (F)	(E)	(Δ)	(c) (F)	(E)	(Δ)	(d) (F)	(E)	(Δ)	(e) (F)	(E)	(Δ)	(f) (F)	(E)	(Δ)	phyto-toxicity corn
(Ib-3)+(B-1) 20 + 250	90	52	38	100	20	80	70	20	50	90	20	70	100	40	60	100	30	70	0
(Ib-3)+(B-2) 20 + 125	100	88	12	100	20	80	100	20	80	100	52	48	100	76	24	80	30	50	0
(Ib-3)+(B-3) 40 + 62	100	95	5	100	93	7	100	68	32	100	50	50	100	76	24	90	40	50	0
(Ib-3)+(B-4) 40 + 125	100	60	40	90	30	60	90	20	70	90	60	30	100	84	16	100	40	60	0
(Ib-3)+(B-5) 20 + 62	90	52	38	90	36	54	40	20	20	90	20	70	90	40	50	90	30	60	0
(Ib-3)+(B-6) 40 + 250	100	60	40	90	30	60	100	36	64	100	50	50	100	60	40	90	40	50	0
(Ib-3)+(B-7) 40 + 125	100	95	5	100	93	7	70	52	18	70	50	20	80	60	20	70	40	30	0
(Ib-3)+(B-8) 20 + 250	100	95	5	100	30	70	60	20	40	100	50	50	100	60	40	80	40	40	0
(Ib-3)+(B-9) 40 + 250	100	95	5	90	44	46	100	36	64	60	50	10	80	60	20	70	40	30	0
(Ib-3)+(B-10) 40 + 250	100	95	5	100	86	14	80	20	60	90	50	40	90	60	30	80	40	40	0
(Ib-3)+(B-14) 40 + 16	60	60	0	50	44	6	40	36	4	80	80	0	80	80	0	80	76	4	0
(Ib-3)+(B-15) 40 + 10	70	60	10	50	44	6	40	36	4	80	75	5	90	80	10	80	70	10	0
(Ib-3)+(B-16) 40 + 36	70	50	20	50	30	20	50	36	14	90	90	0	90	88	2	90	88	2	0
(Ib-3)+(B-17) 40 + 16	80	65	15	50	44	6	40	36	4	70	60	10	70	68	2	50	46	4	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass
Difference (Δ) = Found value(F) - Expected value(E)
A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 10, 11 and 12 show that all the compositions containing Compound one of Compounds (Ib-1), (Ib-2) and (Ib-3) and one of compounds (B-1) ~ (B-10) and (B-14) ~ (B-17) in combination showed high synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing Compound (Ib-1) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ib-1) and Compound (B-1) showed a high synergistic effect on velvet leaf, green foxtail, crabgrass and barnyard grass in particular,

the composition containing Compound (Ib-1) and Compound (B-2) or (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ib-1) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,

the composition containing Compound (Ib-1) and Compound (B-5) showed a high synergistic effect on velvet leaf, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-1) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-1) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-1) and Compound (B-8) showed a high synergistic effect on velvet leaf and green foxtail in particular,
the composition containing Compound (Ib-1) and Compound (B-9) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-1) and Compound (B-10) showed a high synergistic effect on green foxtail in particular, and
the composition containing Compound (Ib-1) and Compound (B-14) or (B-16) showed a high synergistic effect on velvet leaf in particular.

In the compositions containing Compound (Ib-2) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Composition (Ib-2) and Compound (B-1) showed a high synergistic effect on velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-2) or (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-5) showed a high synergistic effect on cocklebur, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-7) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-2) and Compound (B-8) showed a high synergistic effect on velvet leaf, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-9) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-2) and Compound (B-10) showed a high synergistic effect on green foxtail and barnyard grass in particular, and
the composition containing Compound (Ib-2) and Compound (B-15) showed a high synergistic effect on slender amaranth in particular.

In the compositions containing Compound (Ib-3) and one of Compounds (B-1) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ib-3) and compound (B-1) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-2) or (B-4) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-5) showed a high synergistic effect on velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-6) showed a high synergistic effect on velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-7) showed a high synergistic effect on barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-8) showed a high synergistic effect on velvet leaf and green foxtail in particular,
the composition containing Compound (Ib-3) and Compound (B-9) or (B-10) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ib-3) and Compound (B-16) showed a high synergistic effect on cocklebur and velvet leaf in particular, and
the composition containing Compound (Ib-3) and Compound (B-17) showed a high synergistic effect on cocklebur

in particular.

Further, the compositions containing Compound (Ib-1), (Ib-2) or (Ib-3) and one of Compounds (B-1) ~ (B-6) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

Table 13 shows the results of foliar treatment tests on single active ingredients of Compound (Ic-1) and Compounds (B-1) ~ (B-4), (B-6) ~ (B-10) and (b-14) ~ (B-17).

Table 14 shows the results of foliar treatment tests on compositions of Compound (Ic-1) and one of Compounds (B-1) ~ (B-4), (B-6) ~ (B-10) and (b-14) ~ (B-17).

Table 13

Foliar treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ic-1)	40	20	20	0	20	20	20	0
	20	0	0	0	0	0	0	0
(B-1)	250	0	20	40	0	20	0	0
(B-2)	250	0	0	0	20	80	0	0
(B-3)	125	80	80	80	80	20	80	0
(B-4)	500	80	90	60	40	60	0	0
(B-6)	250	0	0	0	0	0	0	0
(B-7)	125	80	90	40	0	0	0	0
(B-8)	125	90	0	0	0	0	0	0
(B-9)	125	90	0	40	0	0	0	0
(B-10)	125	20	10	0	0	20	0	0
(B-14)	16	20	20	20	60	50	60	0
(B-15)	10	10	10	20	50	60	50	0
(B-16)	36	0	0	20	80	70	80	0
(B-17)	16	30	20	20	20	20	10	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Table 14 Foliar treatment (combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%) (a) (F)	(a) (E)	(a) (Δ)	(b) (F)	(b) (E)	(b) (Δ)	(c) (F)	(c) (E)	(c) (Δ)	(d) (F)	(d) (E)	(d) (Δ)	(e) (F)	(e) (E)	(e) (Δ)	(f) (F)	(f) (E)	(f) (Δ)	phyto-toxicity corn
(Ic-1)+(B-1) 40 + 250	100	20	80	100	20	80	60	40	20	40	20	20	100	36	64	30	20	10	0
(Ic-1)+(B-2) 40 + 250	100	20	80	100	20	80	20	0	20	80	36	44	100	80	20	80	20	60	0
(Ic-1)+(B-3) 40 + 125	100	84	16	100	84	16	100	80	20	80	20	60	40	20	20	80	20	60	0
(Ic-1)+(B-4) 40 + 500	100	84	16	100	92	8	100	60	40	90	52	38	100	68	32	60	20	40	0
(Ic-1)+(B-6) 40 + 250	100	20	80	100	20	80	20	0	20	20	20	0	100	20	80	20	20	0	0
(Ic-1)+(B-7) 40 + 125	100	84	16	100	92	8	100	40	60	50	20	30	20	20	0	20	20	0	0
(Ic-1)+(B-8) 40 + 125	100	92	8	20	20	0	100	0	100	60	20	40	60	20	40	40	20	20	0
(Ic-1)+(B-9) 20 + 125	100	90	10	40	0	40	100	40	60	0	0	0	20	0	20	0	0	0	0
(Ic-1)+(B-10) 40 + 125	100	36	64	100	28	72	20	0	20	60	20	40	80	36	44	60	20	40	0
(Ic-1)+(B-14) 40 + 16	40	36	4	40	36	4	40	20	20	70	68	2	80	68	20	70	68	2	0
(Ic-1)+(B-15) 40 + 10	60	27	33	50	27	23	40	20	20	70	60	10	80	62	18	80	60	20	0
(Ic-1)+(B-16) 40 + 36	60	20	40	40	20	20	50	20	30	90	84	6	90	76	14	90	84	6	0
(Ic-1)+(B-17) 40 + 16	80	44	36	40	36	4	40	20	20	60	36	24	70	36	34	50	28	22	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass
Difference (Δ) = Found value(F) - Expected value(E)
A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

All the compositions containing Compound (Ic-1) and one of Compounds (B-1) ~ (B-4), (B-6) ~ (B-10) and (B-14) ~ (B-17) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing Compound (Ic-1) and one of Compounds B-1) ~ (B-4), (B-6) ~ (B-10) and (B-14) ~ (B-17),

the composition containing Compound (Ic-1) and Compound (B-1) or (B-6) showed a high synergistic effect on cocklebur, velvet leaf and crabgrass in particular,

the composition containing Compound (Ic-1) and Compound (B-) showed a high synergistic effect on in particular,

the composition containing Compound (Ic-1) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-3) showed a high synergistic effect on green foxtail

and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-4) showed a high synergistic effect on slender amaranth, green foxtail and barnyard grass in particular,

the composition containing Compound (Ic-1) and one of Compounds (B-7) ~ (B-9) showed a high synergistic effect on slender amaranth in particular,

the composition containing Compound (Ic-1) and Compound (B-10) showed a high synergistic effect on cocklebur and velvet leaf in particular,

the composition containing Compound (Ic-1) and Compound (B-14) showed a high synergistic effect on slender amaranth and crabgrass in particular,

the composition containing Compound (Ic-1) and Compound (B-15) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and barnyard grass in particular,

the composition containing Compound (Ic-1) and Compound (B-16) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and

the composition containing Compound (Ic-1) and Compound (B-17) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail, crabgrass and barnyard grass in particular.

Further, the compositions containing Compound (Ic-1) and one of Compounds (B-1) ~ (B-4), (B-6) and (B-10) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

Example 2 [Soil treatment test]

Seeds of weeds such as cocklebur, velvet leaf, Slender amaranth, green foxtail, crabgrass and barnyard grass and seeds of corn were sown in 1/2,000-are Wagner pots filled with upland soil, and covered with upland soil. One day after the seeds were sown, a predetermined amount of the herbicide obtained in the same manner as in Preparation Example 5 except that one of Compounds (Ia-2) ~ (Ia-5), (Ia-7), (Ib-1) ~ (Ib-3), (Ic-1) and (Ic-2) was suspended in water and uniformly sprayed in a solution amount of 1,000 liters/hectare.

Twenty days after the treatment, the herbicide was determined for phytotoxicity to crops and herbicidal efficacy in the same manner as in the foliar treatment test. The herbicidal efficacy (weed control ratio %) was determined on the basis of the above equation (A).

Table 15 shows the results of soil treatment tests on single active ingredients of Compounds (Ia-2) ~ (Ia-5) and Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 16 to 20 show the results of soil treatment tests on compositions of one of Compounds (Ia-2) ~ (Ia-5) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 15

Soil treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ia-2)	80	20	80	20	20	20	40	0
	40	0	60	0	10	20	40	0
(Ia-3)	80	40	20	0	40	20	40	0
	40	20	20	0	10	20	30	0
(Ia-4)	80	40	60	0	40	50	30	0
	40	20	30	0	20	20	20	0
(Ia-5)	80	40	60	0	30	50	40	0
	40	20	20	0	20	30	20	0
(Ia-7)	80	40	50	40	40	50	40	0
	40	20	30	10	20	30	30	0
(B-7)	125	90	40	0	0	0	0	0
(B-3)	125	90	90	90	20	0	20	0
(B-2)	125	20	0	0	0	0	0	0
(B-4)	500	0	20	20	0	0	0	0
(B-12)	500	20	0	0	60	60	80	0
(B-13)	250	0	20	20	80	80	80	0
(B-11)	250	0	0	0	60	80	40	0
(B-18)	250	20	10	40	70	70	70	0
(B-19)	250	0	0	10	50	70	60	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Table 16 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-2)+(B-7) 40 + 125	100	90	10	100	76	24	100	0	100	20	10	10	100	20	80	80	20	60	0
(Ia-2)+(B-3) 40 + 125	100	90	10	100	95	5	100	90	10	100	36	64	100	20	80	100	52	48	0
(Ia-2)+(B-2) 80 + 125	100	36	64	100	80	20	100	20	80	40	20	20	100	20	80	100	40	60	0
(Ia-2)+(B-4) 40 + 500	90	20	70	100	84	16	100	20	80	40	20	20	60	20	40	100	40	60	0
(Ia-2)+(B-12) 80 + 500	80	36	44	100	20	80	100	20	80	90	68	22	90	68	22	100	98	2	0
(Ia-2)+(B-13) 80 + 250	20	20	0	100	84	16	100	36	64	20	10	10	100	20	80	80	40	40	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 17 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-3)+(B-7) 40 + 125	100	92	8	100	52	48	40	0	40	20	10	10	60	20	40	60	30	30	0
(Ia-3)+(B-3) 40 + 125	100	92	8	100	92	8	100	90	10	90	28	62	100	20	80	100	44	56	0
(Ia-3)+(B-2) 80 + 125	100	52	48	100	20	80	80	0	80	80	40	40	90	20	70	90	40	50	0
(Ia-3)+(B-4) 40 + 500	80	20	60	90	36	54	100	20	80	80	10	70	90	20	70	100	30	70	0
(Ia-3)+(B-12) 80 + 500	80	52	28	80	20	60	60	0	60	90	76	14	100	68	32	100	94	6	0
(Ia-3)+(B-13) 80 + 250	50	40	10	80	36	44	80	20	60	100	88	12	100	84	16	100	94	6	0
(Ia-3)+(B-11) 80 + 250	60	40	20	90	20	70	80	0	80	100	76	24	100	84	16	100	64	36	0
(Ia-3)+(B-18) 80 + 250	80	52	28	80	28	52	80	40	40	100	82	18	100	76	24	100	82	18	0
(Ia-3)+(B-19) 80 + 250	80	40	40	80	20	60	90	10	80	100	70	30	100	76	24	100	76	24	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 18 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-4)+(B-7) 40 + 125	100	92	8	100	58	42	40	0	40	40	20	20	50	20	30	50	20	30	0
(Ia-4)+(B-3) 40 + 125	100	92	8	100	93	7	100	90	10	80	36	44	100	20	80	100	36	64	0
(Ia-4)+(B-2) 80 + 125	100	52	48	100	60	40	70	0	70	80	40	40	100	50	50	90	30	60	0
(Ia-4)+(B-4) 40 + 500	90	20	70	90	44	46	100	20	80	90	20	70	90	20	70	100	20	80	0
(Ia-4)+(B-12) 80 + 500	90	52	38	90	60	30	60	0	60	90	76	14	100	80	20	100	93	7	0
(Ia-4)+(B-13) 80 + 250	50	40	10	80	68	12	60	20	40	100	88	12	100	90	10	100	93	7	0
(Ia-4)+(B-11) 80 + 250	70	30	40	100	60	40	60	0	60	100	76	24	100	90	10	100	58	42	0
(Ia-4)+(B-18) 80 + 250	90	52	38	100	64	36	80	40	40	100	82	18	100	85	15	100	79	21	0
(Ia-4)+(B-19) 80 + 250	90	40	50	80	60	20	100	10	90	100	70	30	100	85	15	100	72	28	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 19 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ia-5)+(B-7) 40 + 125	100	92	8	100	52	48	70	0	70	40	20	20	50	30	20	40	20	20	0
(Ia-5)+(B-3) 40 + 125	100	92	8	100	92	8	90	90	10	80	36	44	100	30	70	100	36	64	0
(Ia-5)+(B-2) 80 + 125	100	52	48	100	60	40	80	60	20	90	30	60	100	50	50	90	40	50	0
(Ia-5)+(B-4) 40 + 500	100	20	80	100	36	64	100	20	80	90	20	70	90	30	60	100	20	80	0
(Ia-5)+(B-12) 80 + 500	100	52	48	100	60	40	60	0	60	90	72	18	100	80	20	100	94	6	0
(Ia-5)+(B-13) 80 + 250	50	40	10	70	68	2	40	20	20	100	86	14	100	90	10	100	94	6	0
(Ia-5)+(B-11) 80 + 250	90	40	50	100	60	40	40	0	40	100	72	28	100	90	10	100	64	36	0
(Ia-5)+(B-18) 80 + 250	90	52	38	90	64	26	80	40	40	100	79	21	100	85	15	100	82	18	0
(Ia-5)+(B-19) 80 + 250	80	40	40	90	60	30	100	10	90	100	65	35	100	85	15	100	76	24	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 20 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			corn
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	
(Ia-7)+(B-7) 40 + 125	100	92	8	100	58	42	80	10	70	50	20	30	50	30	20	90	20	70	0
(Ia-7)+(B-3) 40 + 125	100	92	8	100	93	7	100	91	9	80	36	44	100	30	70	100	44	56	0
(Ia-7)+(B-2) 80 + 125	100	52	48	100	50	50	80	20	60	90	40	50	100	50	50	100	40	60	0
(Ia-7)+(B-4) 40 + 500	90	20	70	100	44	56	100	36	64	90	20	70	100	30	70	100	30	70	0
(Ia-7)+(B-12) 80 + 500	100	52	48	100	50	50	80	20	60	100	76	24	100	80	20	100	94	6	0
(Ia-7)+(B-13) 80 + 250	60	40	20	80	60	20	80	36	44	100	88	12	100	90	10	100	94	6	0
(Ia-7)+(B-11) 80 + 250	70	40	30	90	50	40	60	20	40	100	76	24	100	90	10	100	64	36	0
(Ia-7)+(B-18) 80 + 250	80	52	28	90	55	35	80	52	28	100	82	18	100	85	15	100	82	18	0
(Ia-7)+(B-19) 80 + 250	100	40	60	90	50	40	100	28	72	100	70	30	100	85	15	100	76	24	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 16 ~ 20 show that all the compositions containing Compound (Ia-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-12) and (B-13) in combination and all the compositions containing one of Compounds (Ia-2) ~ (Ia-5) and (Ia-7) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing (Ia-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-12) and (B-13),

the composition containing Compound (Ia-2) and Compound (B-7) showed a high synergistic effect on crabgrass, barnyard grass and slender amaranth in particular,
the composition containing Compound (Ia-2) and Compound (B-) showed a high synergistic effect on in particular,
the composition containing Compound (Ia-2) and Compound (B-3) showed a high synergistic effect on green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-2) and Compound (B-2) showed a high synergistic effect on cocklebur, slender amaranth, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-2) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-2) and Compound (B-12) showed a high synergistic effect on velvet leaf and slender amaranth in particular, and
the composition containing Compound (Ia-2) and Compound (B-13) showed a high synergistic effect on slender amaranth and crabgrass in particular.

Further, the compositions containing Compound (Ia-2) and one of Compounds (B-2) ~ (B-4) and (B-7) showed a herbicidal effect at an earlier stage than any individual herbicide used as a single active ingredient.

In the compositions containing Compound (Ia-3) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-3) and Compound (B-7) showed a high synergistic effect on velvet leaf, slender amaranth and crabgrass in particular,
the composition containing Compound (Ia-3) and Compound (B-3) showed a high synergistic effect on green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-3) and Compound (B-12) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-3) and Compound (B-13) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-3) and Compound (B-11) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-3) and Compound (B-18) showed a high synergistic effect on velvet leaf and slender amaranth in particular, and
the composition containing Compound (Ia-3) and Compound (B-19) showed a high synergistic effect on velvet leaf and slender amaranth in particular.

In the compositions containing Compound (Ia-4) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-4) and Compound (B-7) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-4) and Compound (B-3) showed a high synergistic effect on green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-2) showed a high synergistic effect on cocklebur, slender amaranth, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-12) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-4) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-4) and Compound (B-11) showed a high synergistic effect on cocklebur, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-4) and Compound (B-18) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and

the composition containing Compound (Ia-4) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing Compound (Ia-5) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-5) and Compound (B-7) showed a high synergistic effect on velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-5) and Compound (B-3) showed a high synergistic effect on crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-2) showed a high synergistic effect on cocklebur, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-12) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-5) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-5) and Compound (B-11) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-5) and Compound (B-18) showed a high synergistic effect on cocklebur and slender amaranth in particular, and
the composition containing Compound (Ia-5) and Compound (B-19) showed a high synergistic effect on slender amaranth in particular.

In the compositions containing Compound (Ia-7) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ia-7) and Compound (B-7) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-3) showed a high synergistic effect on crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-4) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-12) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,
the composition containing Compound (Ia-7) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular,
the composition containing Compound (Ia-7) and Compound (B-11) showed a high synergistic effect on velvet leaf, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ia-7) and Compound (B-18) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and
the composition containing Compound (Ia-7) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

Table 21 shows the results of soil treatment tests on single active ingredients of Compounds (Ib-1) ~ (Ib-3) and Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 22 to 24 show the results of soil treatment tests on compositions of one of Compounds (Ib-1) ~ (Ib-3) and one of compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 21

Soil treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ib-1)	80	20	60	20	40	80	40	0
	40	0	20	0	20	50	20	0
(Ib-2)	80	10	60	20	30	70	40	0
	40	0	20	0	0	20	0	0
(Ib-3)	80	20	50	20	30	60	40	0
(B-2)	125	0	0	0	0	0	0	0
(B-3)	62	90	90	60	0	40	0	0
(B-4)	250	0	20	20	0	0	0	0
(B-7)	62	40	20	0	0	0	0	0
(B-11)	250	0	0	0	60	80	40	0
(B-12)	125	0	0	0	20	60	60	0
(B-13)	125	0	0	0	80	80	20	0
(B-18)	250	20	10	40	70	70	70	0
(B-19)	250	0	0	10	50	70	60	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Table 22 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ib-1)+(B-2) 80 + 125	80	20	60	100	60	40	100	20	80	100	40	60	100	80	20	100	40	60	0
(Ib-1)+(B-3) 40 + 62	100	90	10	100	92	8	100	60	40	40	20	20	100	70	30	90	20	70	0
(Ib-1)+(B-4) 80 + 250	80	20	60	100	68	32	80	36	44	80	40	40	80	40	40	90	80	10	0
(Ib-1)+(B-7) 80 + 62	100	52	48	100	68	32	20	20	0	80	40	40	90	80	10	80	40	40	0
(Ib-1)+(B-11) 80 + 250	60	20	40	100	60	40	20	20	0	100	76	24	100	96	4	100	64	36	0
(Ib-1)+(B-12) 80 + 250	60	20	40	100	60	40	100	20	80	100	52	48	100	92	8	100	76	24	0
(Ib-1)+(B-13) 80 + 125	40	20	20	100	60	40	100	20	80	100	88	12	100	96	4	100	52	48	0
(Ib-1)+(B-18) 80 + 250	80	36	44	80	64	16	90	52	38	90	82	8	100	94	6	100	82	18	0
(Ib-1)+(B-19) 80 + 250	90	20	70	90	60	30	100	28	72	100	70	30	100	94	6	100	76	24	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 23 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ib-2)+(B-2) 80 + 125	80	10	70	100	60	40	90	20	70	100	30	70	100	70	30	100	40	60	0
(Ib-2)+(B-3) 40 + 62	100	90	10	100	92	8	100	60	40	50	0	50	90	52	38	90	0	90	0
(Ib-2)+(B-4) 80 + 250	80	10	70	100	68	32	90	36	54	90	30	60	90	70	20	90	40	50	0
(Ib-2)+(B-7) 80 + 62	90	46	44	100	68	32	40	20	20	60	30	30	70	70	0	60	40	20	0
(Ib-2)+(B-11) 80 + 250	50	10	40	100	60	40	40	20	20	100	72	28	100	94	6	100	64	36	0
(Ib-2)+(B-12) 8(+ 250	40	10	30	100	60	40	90	20	70	100	44	56	100	88	12	100	76	24	0
(Ib-2)+(B-13) 80 + 125	50	10	40	100	60	40	100	20	80	100	86	14	100	94	6	100	52	48	0
(Ib-2)+(B-18) 80 + 250	80	28	52	90	64	26	100	52	48	100	79	21	100	91	9	100	82	18	0
(Ib-2)+(B-19) 80 + 250	90	10	80	90	60	30	100	28	72	100	65	35	100	91	9	100	76	24	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 24 Soil treatment (combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%) (a) (F)	(a) (E)	(a) (Δ)	(b) (F)	(b) (E)	(b) (Δ)	(c) (F)	(c) (E)	(c) (Δ)	(d) (F)	(d) (E)	(d) (Δ)	(e) (F)	(e) (E)	(e) (Δ)	(f) (F)	(f) (E)	(f) (Δ)	phyto-toxicity corn
(Ib-3)+(B-2) 80 + 125	90	20	70	100	50	50	90	20	70	100	30	70	100	60	40	100	40	60	0
(Ib-3)+(B-3) 40 + 62	100	92	8	100	95	5	90	68	22	80	30	50	100	76	24	90	40	50	0
(Ib-3)+(B-4) 80 + 250	80	20	60	100	60	40	90	36	54	80	30	50	80	60	20	90	40	50	0
(Ib-3)+(B-7) 80 + 62	90	52	38	100	60	40	50	20	30	70	30	40	90	60	30	80	40	40	0
(Ib-3)+(B-11) 80 + 250	60	20	40	100	50	50	60	20	40	100	72	28	100	92	8	100	64	36	0
(Ib-3)+(B-12) 80 + 250	80	20	60	100	50	50	100	20	80	100	44	56	100	84	16	90	76	14	0
(Ib-3)+(B-13) 80 + 125	70	20	50	100	50	50	100	20	80	100	86	14	100	92	8	100	52	48	0
(Ib-3)+(B-18) 80 + 250	80	36	44	100	55	45	100	52	48	90	79	11	100	88	12	100	82	18	0
(Ib-3)+(B-19) 80 + 250	80	20	60	90	50	40	100	28	72	100	65	35	100	88	12	100	76	24	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 22, 23 and 24 show that all the compositions containing one of Compounds (Ib-1), (Ib-2) and (Ib-3) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing (Ib-1) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18)

and (B-19),

the composition containing Compound (Ib-1) and Compound (B-2) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-1) and Compound (B-3) showed a high synergistic effect on barnyard grass in particular,
the composition containing Compound (Ib-1) and Compound (B-4) showed a high synergistic effect on cocklebur in particular,
the composition containing Compound (Ib-1) and Compound (B-7) showed a high synergistic effect on cocklebur in particular,
the composition containing Compound (Ib-1) and Compound (B-11) showed a high synergistic effect on cocklebur, velvet leaf and barnyard grass in particular,
the composition containing Compound (Ib-1) and Compound (B-12) or (B-13) showed a high synergistic effect on slender amaranth in particular, and
the composition containing Compound (Ib-1) and Compound (B-18) or (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing (Ib-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ib-2) and Compound (B-2) or (B-4) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-2) and Compound (B-7) showed a high synergistic effect on cocklebur in particular,
the composition containing Compound (Ib-2) and Compound (B-11) showed a high synergistic effect on cocklebur and velvet leaf in particular,
the composition containing Compound (Ib-2) and Compound (B-12) showed a high synergistic effect on slender amaranth and green foxtail in particular,
the composition containing Compound (Ib-2) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular, and
the composition containing Compound (Ib-2) and Compound (B-18) or (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

In the compositions containing (Ib-3) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ib-3) and Compound (B-2) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-3) showed a high synergistic effect on green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-7) showed a high synergistic effect on velvet leaf, green foxtail and barnyard grass in particular,
the composition containing Compound (Ib-3) and Compound (B-11) showed a high synergistic effect on velvet leaf in particular,
the composition containing Compound (Ib-3) and Compound (B-12) showed a high synergistic effect on cocklebur, velvet leaf, slender amaranth and green foxtail in particular,
the composition containing Compound (Ib-3) and Compound (B-13) or (B-18) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular, and
the composition containing Compound (Ib-3) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

Table 25 shows the results of soil treatment tests on single active ingredients of Compounds (Ic-1) and (Ic-2) and Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 26 ~ 27 show the results of soil treatment tests on compositions of one of Compounds (Ic-1) and (Ic-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19).

Table 25

Soil treatment (single active ingredient)								
Compd	Dosage (g/ha)	herbicidal efficacy %						phytotoxicity to corn
		(a)	(b)	(c)	(d)	(e)	(f)	
(Ic-1)	80	50	60	0	30	40	30	0
(Ic-2)	80	40	60	0	40	40	30	0
	40	0	40	0	0	20	0	0
(B-2)	250	60	0	0	0	20	0	0
(B-3)	125	90	90	90	0	40	40	0
(B-4)	500	0	0	0	0	0	0	0
(B-7)	125	80	20	20	0	0	0	0
(B-11)	250	0	0	0	60	80	80	0
(B-12)	250	0	0	0	60	60	90	0
(B-13)	250	0	0	0	90	90	90	0
(B-18)	250	20	10	40	70	70	70	0
(B-19)	250	0	0	10	50	70	60	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Table 26 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	corn
(Ic-1)+(B-2) 80 + 250	100	80	20	100	60	40	90	0	90	80	30	50	100	52	48	100	30	70	0
(Ic-1)+(B-3) 80 + 125	100	95	5	100	96	4	100	90	10	40	30	10	100	64	36	90	58	32	0
(Ic-1)+(B-4) 80 + 500	90	50	40	100	60	40	100	0	100	60	30	30	60	40	20	100	30	70	0
(Ic-1)+(B-7) 80 + 125	100	90	10	100	68	32	40	20	20	40	30	10	90	40	50	100	30	70	0
(Ic-1)+(B-11) 80 + 250	80	50	30	100	60	40	30	0	30	100	72	28	100	88	12	100	86	14	0
(Ic-1)+(B-12) 80 + 250	60	50	10	100	60	40	100	0	100	80	72	8	100	76	24	100	93	7	0
(Ic-1)+(B-13) 80 + 250	80	50	30	100	60	40	100	0	100	100	93	7	100	90	10	100	90	10	0
(Ic-1)+(B-18) 80 + 250	100	60	40	100	64	36	90	40	50	100	79	21	100	82	18	100	79	21	0
(Ic-1)+(B-19) 80 + 250	90	50	40	100	60	40	80	10	70	100	65	35	100	82	18	100	72	28	0

(a): Cocklebur
(b): Velvet leaf
(c): Slender amaranth
(d): Green foxtail
(e): Crabgrass
(f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Table 27 Soil treatment
(combined use of active ingredients)

Dosage of each active ingredient (g/ha)	herbicidal efficacy (%)																		phyto-toxicity
	(a)			(b)			(c)			(d)			(e)			(f)			corn
	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	(F)	(E)	(Δ)	
(Ic-2)+(B-2) 80 + 250	100	76	24	100	60	40	100	0	100	80	40	40	100	52	48	100	30	70	0
(Ic-2)+(B-3) 80 + 125	100	90	10	100	94	6	100	90	10	40	0	40	100	64	36	90	40	50	0
(Ic-2)+(B-4) 80 + 500	90	40	50	100	60	40	100	0	100	60	40	20	60	40	20	100	30	70	0
(Ic-2)+(B-7) 40 + 125	100	88	12	100	68	32	40	20	20	40	40	0	90	40	50	100	30	70	0
(Ic-2)+(B-11) 80 + 250	80	40	40	100	60	40	0	0	0	100	76	24	100	88	12	100	86	14	0
(Ic-2)+(B-12) 80 + 250	60	40	20	100	60	40	100	0	100	80	76	4	100	76	24	100	93	7	0
(Ic-2)+(B-13) 80 + 250	40	40	0	100	60	40	100	0	100	100	94	6	100	94	6	100	93	7	0
(Ic-2)+(B-18) 80 + 250	90	52	38	90	64	26	80	40	40	100	82	18	100	82	18	100	79	21	0
(Ic-2)+(B-19) 80 + 250	90	40	50	90	60	30	80	10	70	100	70	30	100	82	18	100	72	28	0

(a): Cocklebur (d): Green foxtail
(b): Velvet leaf (e): Crabgrass
(c): Slender amaranth (f): Barnyard grass

Difference (Δ) = Found value(F) - Expected value(E)

A larger difference (Δ) means a larger synergistic effect due to the combined use of active ingredients.

Tables 26 and 27 show that all the compositions containing one of Compounds (Ic-1) and (Ic-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19) in combination showed synergistic herbicidal effects on all the weeds used for the test.

That is, in the compositions containing (Ic-1) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18)

and (B-19),

the composition containing Compound (Ic-1) and Compound (B-2) showed a high synergistic effect on slender amaranth, velvet leaf, green foxtail, crabgrass and barnyard grass in particular,
the composition containing Compound (Ic-1) and Compound (B-) showed a high synergistic effect on in particular,
the composition containing Compound (Ic-1) and Compound (B-3) showed a high synergistic effect on barnyard grass in particular,
the composition containing Compound (Ic-1) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth, velvet leaf and barnyard grass in particular,
the composition containing Compound (Ic-1) and Compound (B-7) showed a high synergistic effect on velvet leaf, crabgrass and barnyard grass in particular,
the composition containing Compound (Ic-1) and Compound (B-11) showed a high synergistic effect on cocklebur, velvet leaf and slender amaranth in particular,
the composition containing Compound (Ic-1) and Compound (B-12) showed a high synergistic effect on slender amaranth, velvet leaf and crabgrass in particular, and
the composition containing Compound (Ic-1) and Compound (B-13), (B-18) or (B-19) showed a high synergistic effect on cocklebur, slender amaranth and velvet leaf in particular.

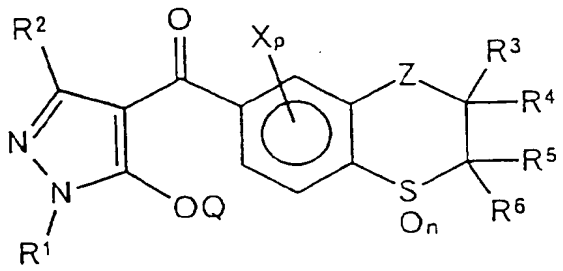
In the compositions containing (Ic-2) and one of Compounds (B-2) ~ (B-4), (B-7), (B-11) ~ (B-13), (B-18) and (B-19),

the composition containing Compound (Ic-2) and Compound (B-2) showed a high synergistic effect on slender amaranth and barnyard grass in particular,
the composition containing Compound (Ic-2) and Compound (B-3) showed a high synergistic effect on barnyard grass in particular,
the composition containing Compound (Ic-2) and Compound (B-4) showed a high synergistic effect on cocklebur, slender amaranth and barnyard grass in particular,
the composition containing Compound (Ic-2) and Compound (B-7) showed a high synergistic effect on crabgrass and barnyard grass in particular,
the composition containing Compound (Ic-2) and compound (B-11) showed a high synergistic effect on cocklebur and velvet leaf in particular,
the composition containing Compound (Ic-2) and Compound (B-12) showed a high synergistic effect on slender amaranth and crabgrass in particular,
the composition containing Compound (Ic-2) and Compound (B-13) showed a high synergistic effect on slender amaranth in particular, and
the composition containing Compound (Ic-2) and Compound (B-19) showed a high synergistic effect on cocklebur and slender amaranth in particular.

The herbicide composition of the present invention not only exhibits high herbicidal efficacy but also has a broad herbicidal spectrum on the basis of the synergistic effect of the pyrazole derivative of the general formula (I) and at least one of Compounds (B-1) ~ (B-20) which are active ingredients thereof. Further, the herbicide composition of the present invention exhibits high activity against weeds which are hard to control. Moreover, the herbicide composition of the present invention has high safety for crops such as corn, etc., and is free from damaging (causing phytotoxicity to) crops.

Claims

1. A herbicide composition containing, as active ingredients, a pyrazole derivative of the general formula (I),



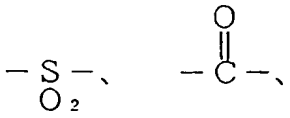
(I)

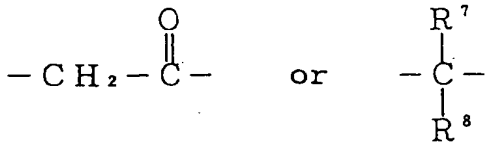
{wherein:

R^1 is a C_1~C_4 alkyl group, a C_2~C_4 alkenyl group or a C_2~C_4 haloalkeny group,
R^2 is a hydrogen atom, a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group or a C_2~C_4 alkoxyalkyl group,
X is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_2-C_4 alkoxyalkyl group, a halogen atom, a C_1~C_4 alkoxy group or a C_1~C_4 haloalkoxy group,
p 15 an integer of 0, 1 or 2,
each of R^3, R^4, R^5 and R^6 is independently a hydrogen atom, a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group or a C_2~C_4 alkoxyalkyl group,
n is an integer of 0, 1 or 2,
Q is a hydrogen atom or a group of A-B,

[in which

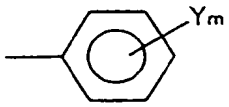
A is a group of





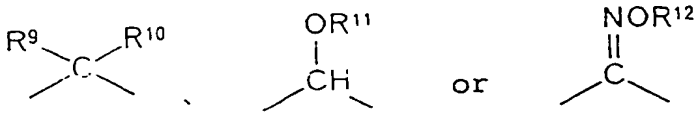
(in which each of R^7 and R^8 is independently a hydrogen atom or a C_1~C_4 alkyl group), and

B is a C_1~C_{12} alkyl group, a C_3~C_{10} cycloalkyl group or a group of



(in which Y is a C_1~C_4 alkyl group, a C_1~C_4 alkoxy group, a C_1~C_4 haloalkyl group, a nitro group or a halogen atom, and

m is an integer of 0 or 1 ~ 3)], and
Z is



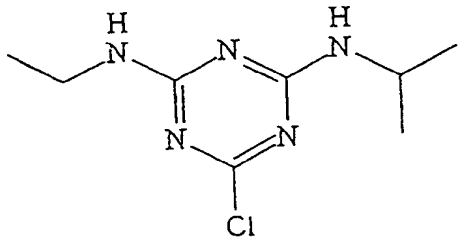
[in which

R^9 is a hydrogen atom, a C_1~C_4 alkyl group or a C_1~C_4 haloalkyl group,
R^{10} is a hydrogen atom, a C_1~C_4 alkyl group, a C_2~C_4 alkenyl group or a C_2~C_4 alkynyl group,
R^{11} is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_3~C_6 cycloalkyl group, a C_3~C_6 alkenylalkyl group, a C_3~C_6 alkynylalkyl group or a C_3~C_6 haloalkenylalkyl group,
R^{12} is a C_1~C_4 alkyl group, a C_1~C_4 haloalkyl group, a C_3~$_6$ cycloalkyl group, a C_3~C_6 alkenylalkyl group, a C_3~C_6 alkynylalkyl group or a C_3~C_6 haloalkenylalkyl group]},

or a salt thereof; and
at least one herbicide compound selected from the group consisting of

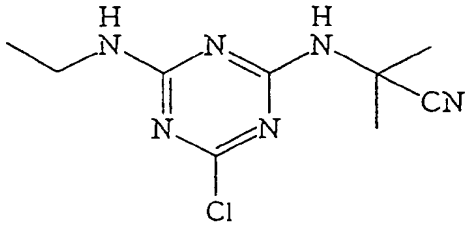
a choroacetamide-based herbicide,
an imidazoline-based herbicide, and
Compound(B-1)

Common name:atrazine
Chemical Name:
6-chloro-N^2-ethyl-N^4-isopropil-1,3,5-triazine-2,4-diamine



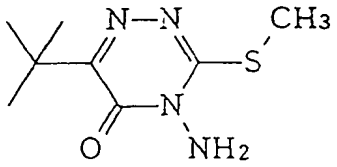
Compound(B-2)

Common name:cyanazine
Chemical Name:
2-(4-chloro-6-ethylamino-1,3,5-triazin-2-ylamino)-2-methylpropionitrile



Compound(B-3)

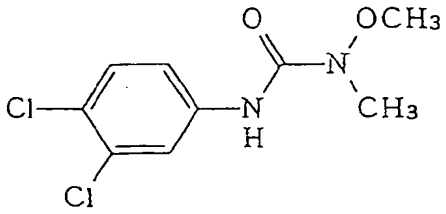
Common name:metribuzin
Chemical Name:
4-amino-6-tert-buthyl-4,5-dihydro-3-methylthio-1,2,4-triazin-5-one



Compound(B-4)

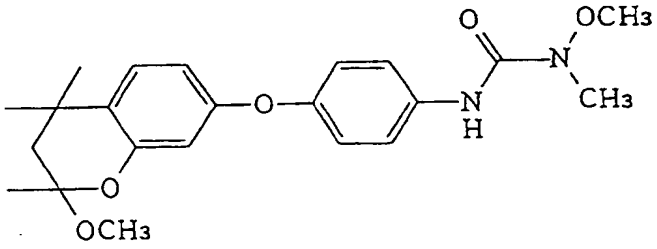
Common name:linuron

Chemical Name:
3-(3,4-dichlorophenyl)-1-methoxy-1-methylurea



Compound(B-5)

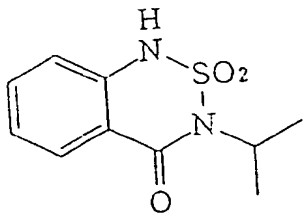
Common name:metbenzurone
Chemical Name:
(±)-1-methoxy-3-[4-(2-methoxy-2,4,4-trimethylchroman-7-yloxy)phenyl]-1-methylurea



Compound(B-6)

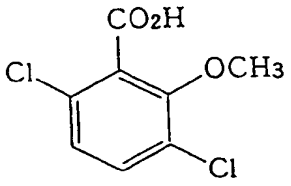
Common name:bentazone

Chemical Name:
3-isopropyl-1H-2,1,3-benzothiadin-4(3H)-one-2,2-dioxide



Compound(B-7)

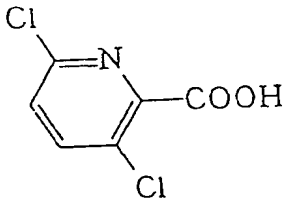
Common name:dicamba
Chemical Name:
3,6-dichloro-2-methoxybenzoic acid



Compound(B-8)

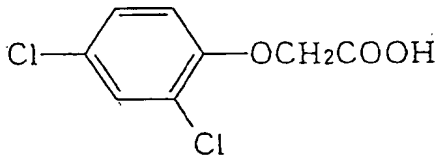
Common name:chlopyralid

Chemical Name:
3,6-dichloropyridine-2-carboxylic acid



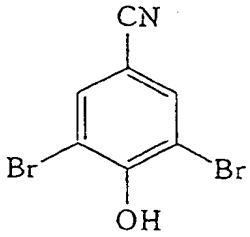
Compound(B-9)

Common name:2,4-D
Chemical Name:
2-(2,4-dichlorophenoxy)acetic acid



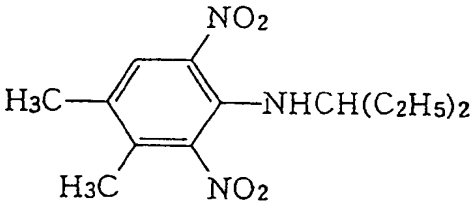
Compound(B-10)

Common name:bromoxynil
Chemical Name:
3,5-dlbromo-4-hydroxybenzonitrile



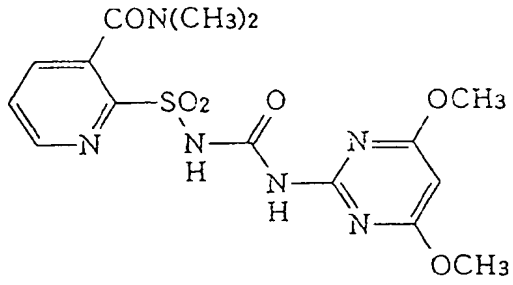
Compound(B-13)

Common name:pendimethalin
Chemical Name:
N-(1-ethylpropyl)-2,6-dinitro-3,4-xylidine



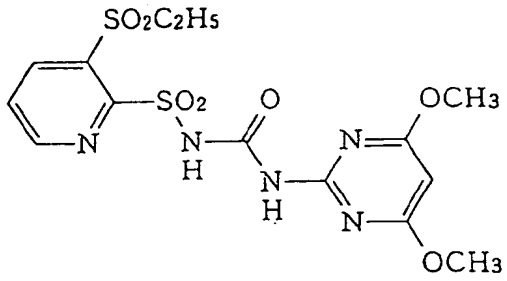
Compound(B-14)

Common name:nicosulfuron
Chemical Name:
2-(4,6-dimethoxypyrimidin-2-ylcarbamoylsulfamoyl)-N,N-dimethylnicotineamide



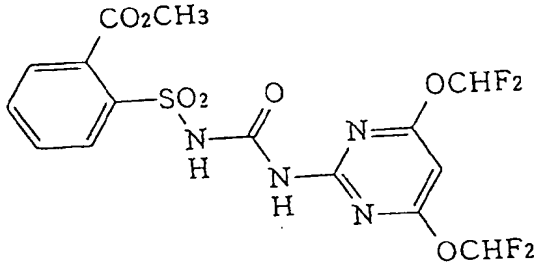
Compound(B-15)

Common name:rimsulfuron
Chemical Name:
1-(4,6-dimethoxypyrimidin-3-(3-ethylsulfonyl-2-pyridylsulfonyl)urea



Compound(B-17)

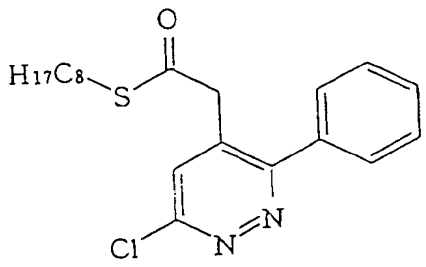
Common name:primisulfuron
Chemical Name:
Methyl 2-[4,6-bis(difluoromethoxy)pyrimidin-2-yl-carbamoylsulfamoyl]benzoic acid



Compound(B-20)

Common name:pyridate
Chemical Name:

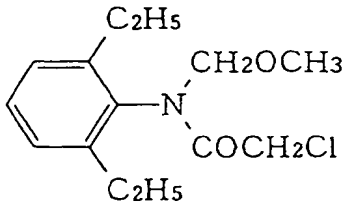
6-chloro-3-phenylpyridazin-4-yl-S-octylthiocarbonate



2. The herbicide composition of Claim 1, wherein the chloroacetamide-based herbicide is at least one herbicide compound selected from the group consisting of

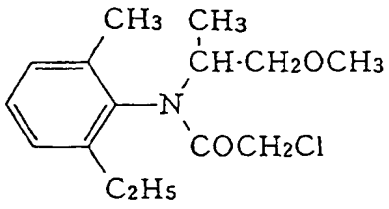
Compound(B-11)

Common name:alachlor
Chemical Name:
2-chloro-2',6'-diethyl-N-methoxymethylacetanilide



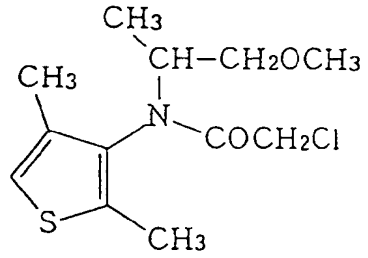
Compound(B-12)

Common name:metolachlor
Chemical Name:
2-chloro-N-(2-ethyl-6-methylphenyl)-N-(2-methoxy-1-methylethyl)acetamide



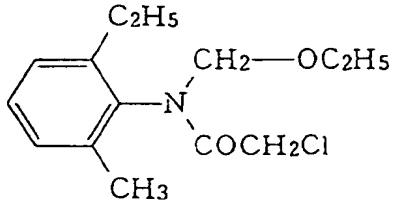
Compound(B-18)

Common name:dimethenamid
Chemical Name:
(1RS,aRS)-2-chloro-N-(2,4-dimethyl-3-thienyl)-N-(2-methoxy-1-methylethyl)acetamide



Compound(B-19)

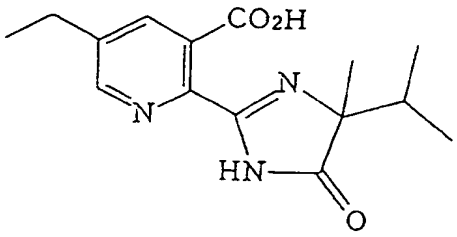
Common name:acetochlor
Chemical Name:
2-chloro-2'-ethyl-6'-methyl-N-ethoxymethylacetanilide



3. The herbicide composition of Claim 1, wherein the imidazoline-based herbicide is at least one herbicide compound selected from the group consisting of

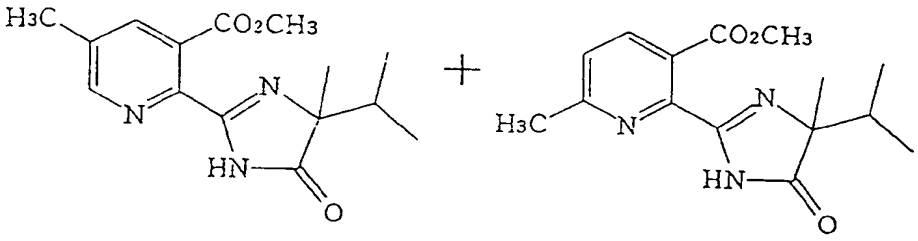
Compound (B-16)

Common name:imazethapyr
Chemical Name:
5-ethyl-2-(4-isopropil-4-methyl-5-oxo-2-imidazolin-2-yl)-nicotinic acid



Compound (B-21)

Common name:imazamethabenz-methyl
Chemical Name:
Compoud of a Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-m-toluic acid and Methyl 6-(4-isopropyl-4-methyl-5-oxo-2-imidazolin-2-yl)-p-toluic acid.



4. The herbicide composition of Claim 1, wherein in the general formula (I) R^1 is a C_1~C_4 alkyl group.

5. The herbicide composition of Claim 1, wherein in the general formula (I) R^2 is a hydrogen atom or a C_1~C_4 alkyl group.

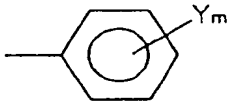
6. The herbicide composition of Claim 1, wherein in the general formula (I) X is a C_1~C_4 alkyl group or a halogen atom.

7. The herbicide composition of Claim 1, wherein substituent(s) X is/are substituted on the 5-position and/or 8-position of a thiochroman ring.

8. The herbicide composition of Claim 1, wherein in the general formula (I) each of R^3, R^4, R^5 and R^6 is independently a hydrogen atom or a C_1~C_4 alkyl group.

9. The herbicide composition of Claim 1, wherein in the general formula (I) n is 2.

10. The herbicide composition of claim 1, wherein in the general formula (I) Q is a group of -A-B in which B is a group of



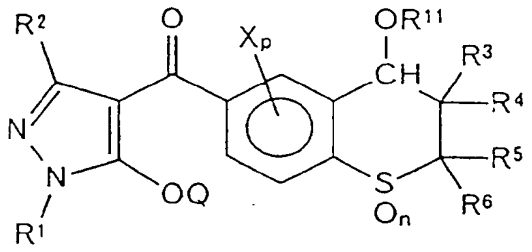
in which Y is a C_1~C_4 alkyl group, a C_1~C_4 alkoxy group, a nitro group or a halogen atom.

11. The herbicide composition of any one of Claims 1 to 3, wherein the herbicide composition contains the pyrazole derivative of the general formula (I) and one of Compounds (B-1) ~ (B-20) in the following mixing ratio (weight ratio),

pyrazole derivative(I):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(I):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(I):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(I):compound(B-4:linuron)=2:1~1:50
pyrazole derivative(I):compound(B-5:metbenzurone)=1:2~ 1:100
pyrazole derivative(I):compound(B-6:bentazone)=6:1~1:100
pyrazole derivative(I):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(I):compound(B-8:chlopyralid)=4:3~1:12
pyrazole derivative(I):compound(B-9:2,4-D)=2:1~1:5
pyrazole derivative(I):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(I):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(I):compound(B-12:metolachlor)=2:1~1:25
pyrazole derivative(I):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(I):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(I):compound(B-15:rimsulfuron)=1:3~40:1

pyrazole derivative(I):compound(B-16:imazethapyr)=1:6~40:1
pyrazole derivative(I):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(I):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(I):compound(B-19:acetochlor)=2:1~1:50
pyrazole derivative(I):compound(B-20:pyridate)=3:2~1:50
pyrazole derivative(I):compound(B-21:imazamethabenzmethyl)=1:6~40:1

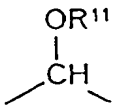
12. The herbicide composition of Claim 1, wherein the pyrazole derivative of the general formula (I) is a pyrazole derivative of the general formula (Ia),



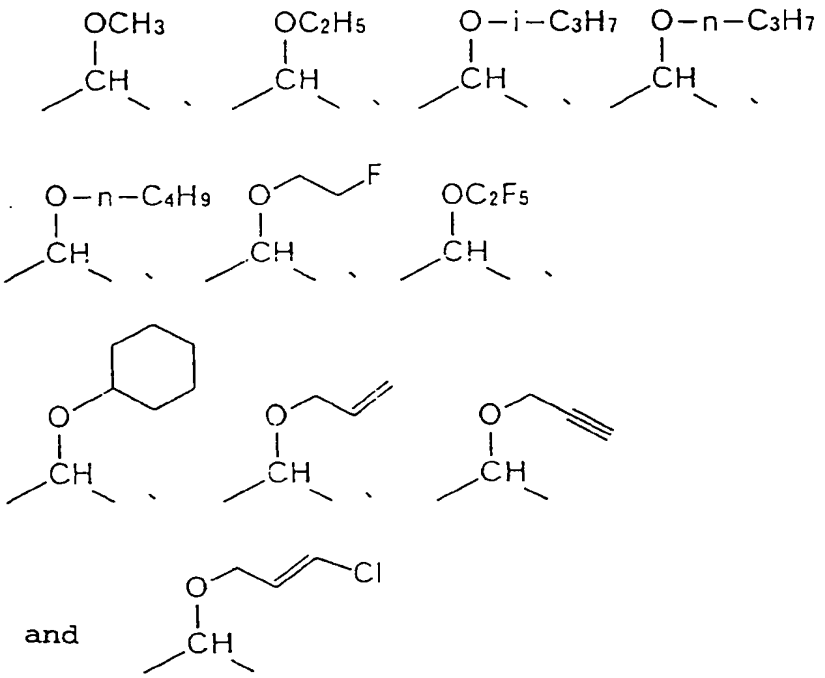
(Ia)

[wherein R^1, R^2, R^3, R^4, R^5, R^6, X, p, n, Q and R^{11} are as defined in Claim 1]
or a salt thereof.

13. The herbicide composition of Claim 12, wherein in the general formula (Ia),



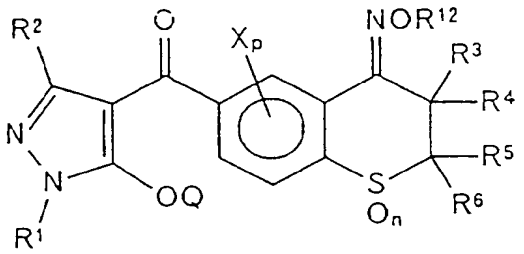
is selected from the group consisting of



14. The herbicide composition of Claim 12, wherein the herbicide composition contains the pyrazole derivative of the general formula (Ia) and one of Compounds (B-1) ~ (B-20) in the following mixing ratio (weight ratio),

pyrazole derivative(Ia):compound(B-1:atrazine)=1:1~1:50
pyrazole derivative(Ia):compound(B-2:cyanazine)=2:1~1:25
pyrazole derivative(Ia):compound(B-3:metribuzin)=3:1~1:12
pyrazole derivative(Ia):compound(B-4:linuron)=2:1~1:25
pyrazole derivative(Ia):compound(B-6:bentazone)=1:21~1:100
pyrazole derivative(Ia):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(Ia):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(Ia):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(Ia):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(Ia):compound(B-12:metolachlor)=2:1~ 1:25
pyrazole derivative(Ia):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(Ia):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(Ia):compound(B-15:rimsulfuron)=1:3~ 40:1
pyrazole derivative(Ia):compound(B-16:imazethapyr)=1:6~ 40:1
pyrazole derivative(Ia):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(Ia):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(Ia):compound(B-19:acetochlor)=2:1~1:50
pyrazole derivative(Ia):compound(B-20:pyridate)=3:2~1:50

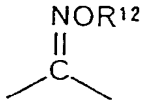
15. The herbicide composition of Claim 1, wherein the pyrazole derivative of the general formula (I) is a pyrazole derivative of the general formula (Ib),



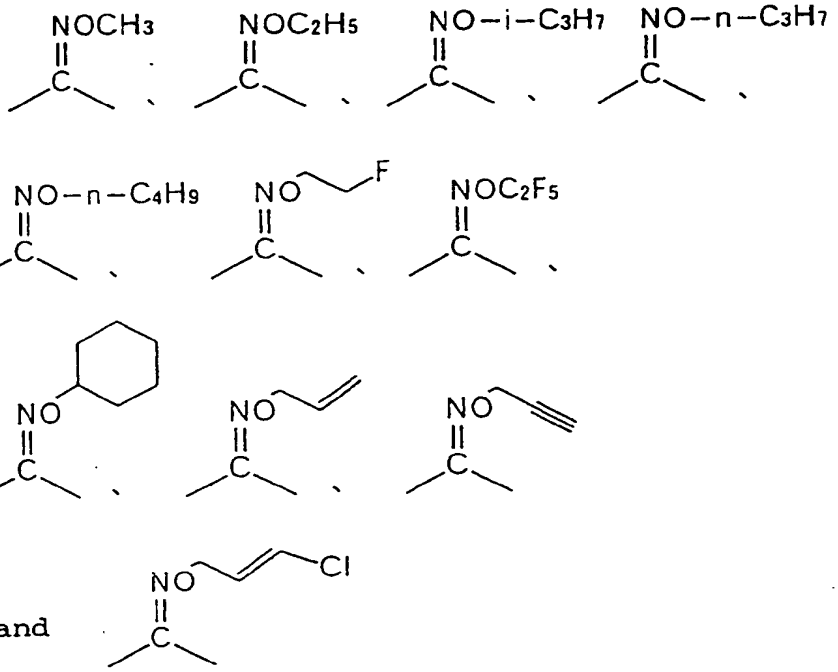
(Ib)

[wherein R^1, R^2, R^3, R^4, R^5, R^6, X, p, n, Q and R^{12} are as defined in Claim 1]
or a salt thereof.

16. The herbicide composition of Claim 15, wherein in the general formula (Ib),



is selected from the group consisting of

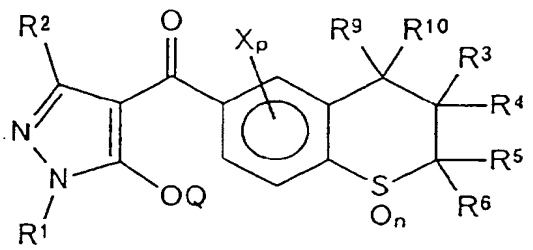


17. The herbicide composition of Claim 15, wherein the herbicide composition contains the pyrazole derivative of the general formula (Ib) and one of Compounds (B-1) ~ (B-19) in the following mixing ratio (weight ratio),

pyrazole derivative(Ib):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(Ib):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(Ib):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(Ib):compound(B-4:linuron)=2:1~1:50

pyrazole derivative(Ib):compound(B-5:metbenzurone)=1:2~ 1:100
pyrazole derivative(Ib):compound(B-6:bentazone)=6:1~1:3
pyrazole derivative(Ib):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(Ib):compound(B-8:chlopyralid)=4:3~1:12
pyrazole derivative(Ib):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(Ib):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(Ib):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(Ib):compound(B-12:metolachlor)=2:1~ 1:25
pyrazole derivative(Ib):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(Ib):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(Ib):compound(B-15:rimsulfuron)=1:3~ 40:1
pyrazole derivative(Ib):compound(B-16:imazethapyr)=1:6~ 40:1
pyrazole derivative(Ib):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(Ib):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(Ib):compound(B-19:acetochlor)=2:1~1:50

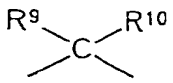
18. The herbicide composition of Claim 1, wherein the pyrazole derivative of the general formula (I) is a pyrazole derivative of the general formula (Ic),



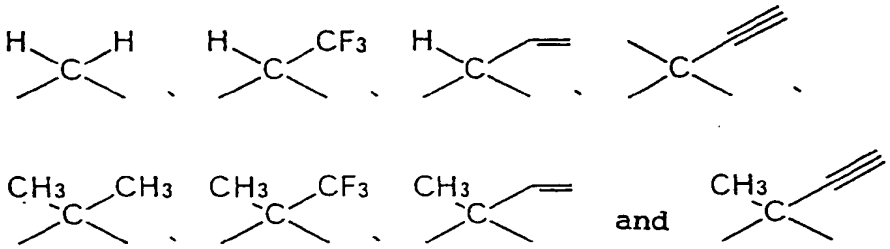
(Ic)

[wherein R^1, R^2, R^3, R^4, R^5, R^6, X, p, n, Q, R^9 and R^{10} are as defined in Claim 1]
or a salt thereof.

19. The herbicide composition of Claim 18, wherein in the general formula (Ic),



is selected from the group consisting of



20. The herbicide composition of Claim 18, wherein the herbicide composition contains the pyrazole derivative of the general formula (Ic) and one of Compounds (B-1) ~ (B-4) and (B-6) ~ (B-19) in the following mixing ratio (weight ratio),

pyrazole derivative(Ic):compound(B-1:atrazine)=2:1~1:50
pyrazole derivative(Ic):compound(B-2:cyanazine)=2:1~1:50
pyrazole derivative(Ic):compound(B-3:metribuzin)=3:1~1:25
pyrazole derivative(Ic):compound(B-4:linuron)=2:1~1:50
pyrazole derivative(Ic):compound(B-6:bentazone)=1:21~1:100
pyrazole derivative(Ic):compound(B-7:dicamba)=1:1~1:50
pyrazole derivative(Ic):compound(B-8:chlopyralid)=4:3~1:12
pyrazole derivative(Ic):compound(B-9:2,4-D)=2:1~1:50
pyrazole derivative(Ic):compound(B-10:bromoxynil)=1:1~1:50
pyrazole derivative(Ic):compound(B-11:alachlor)=2:1~1:25
pyrazole derivative(Ic):compound(B-12:metolachlor)=2:1~ 1:25
pyrazole derivative(Ic):compound(B-13:pendimethalin)=2:1~ 1:25
pyrazole derivative(Ic):compound(B-14:nicosulfuron)=1:3~ 40:1
pyrazole derivative(Ic):compound(B-15:rimsulfuron)=1:3~ 40:1
pyrazole derivative(Ic):compound(B-16:imazethapyr)=1:6~ 40:1
pyrazole derivative(Ic):compound(B-17:primisulfuron)=1:3~ 40:1
pyrazole derivative(Ic):compound(B-18:dimethenamid)=2:1~ 1:50
pyrazole derivative(Ic):compound(B-19:acetochlor)=2:1~1:50

INTERNATIONAL SEARCH REPORT

International application No.
PCT/JP95/01280

A. CLASSIFICATION OF SUBJECT MATTER Int. Cl^6 A01N43/50, A01N43/70, A01N43/707, A01N47/30, A01N43/88, A01N37/40, A01N43/40, A01N37/38, A01N37/34, A01N33/18, A01N47/36, A01N43/58, A01N37/36, A01N43/10
According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols) Int. Cl^6 A01N43/50, A01N43/70, A01N43/707, A01N47/30, A01N43/88, A01N37/40, A01N43/40, A01N37/38, A01N37/34, A01N33/18, A01N47/36, A01N43/58, A01N37/36, A01N43/10

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
CAS ONLINE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	WO, 94/1431, A1 (Idemitsu Kosan Co., Ltd.), January 20, 1994 (20. 01. 94) & JP, 6-503159, A & CN, 1084170, A	1 - 20
A	WO, 93/18031, A1 (Idemitsu Kosan Co., Ltd.), September 16, 1993 (16. 09. 93) & JP, 5-515530, A & EP, 629623, A1	1 - 20

☐ Further documents are listed in the continuation of Box C. ☐ See patent family annex.

* Special categories of cited documents:
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"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art
"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
July 18, 1995 (18. 07. 95)	August 8, 1995 (08. 08. 95)
Name and mailing address of the ISA/ Japanese Patent Office	Authorized officer
Facsimile No.	Telephone No.

Form PCT/ISA/210 (second sheet) (July 1992)